

A MICROSCOPIC AND THERMAL
STUDY OF THE SOLIDIFICATION
OF HEXADECANE

J. O. GOLDEN

F. J. STERMOLE

CHEMICAL AND PETROLEUM REFINING ENGINEERING DEPARTMENT

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OF THE SOLIDIFICATION OF HEXADECANE

by

J. O. Golden
F. J. Stermole

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George C. Marshall Space Flight Center
Marshall Space Flight Center, Alabama 35812

PREFACE

This report was prepared by Colorado School of Mines, Golden, Colorado, under Contract NAS 8-30511 "Research in Phase Change Thermal Control Technology" and under Colorado School of Mines Foundation Contracts F-6911 and F-6915. The work was administered under the direction of the Space Sciences Laboratory, George C. Marshall Space Flight Center, with Mr. T. C. Bannister acting as the contracting officers' technical representative.

This report covers work from 21 November 1968 to 31 December 1969.

The work at Colorado School of Mines was carried out by Dr. J. O. Golden and F. J. Stermole.

ABSTRACT

The recent interest in the n-paraffins as potential candidate phase change thermal control materials necessitates a detailed understanding of their solidification processes both on earth and in space. Therefore, the goal of this investigation is two-fold:

- (1) Develop an improved microphotography apparatus for observing the phase change process
- (2) Perform meaningful studies on the crystallization dynamics of the solidification of the n-paraffins.

The microphotography apparatus was successfully developed with the improved features being variable magnification and a modified test cell mounting procedure. This equipment was then used to study the solidification process of hexadecane under a variety of experimental conditions. Parameters investigated were average interfacial velocity, crystal peak height, individual crystal growth rates, crystal morphology and temperature response of the test cell.

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1.0 INTRODUCTION

In recent years the concept of using the heat associated with the solid-liquid phase transition of a number of materials has received increasing attention as an attractive thermal control concept. This concept involves the idea of using the heat associated with the melting process of a given material to absorb the excess thermal energy of a thermally sensitive component. With a properly designed device, the component can be held below a prescribed temperature bound. Conversely, the component temperature could be held above a prescribed temperature bound by using the heat of fusion of a given material to release energy to the component.

This concept, phase change thermal control, is particularly attractive for use in spacecraft thermal control problems from the standpoint that it is a passive system (requires no moving parts) and, therefore, should have a high reliability. However, a number of points require further investigation before phase change thermal control can be applied to spacecraft problems. One point in question is: what effect does the space environment have on the melting or solidification of the phase change material?

In attempting to answer the previous question, the problem of the mechanism of both melting and solidification of the candidate phase change materials immediately arises. Although a great deal of work has been done in these areas,

crystal growth technology remains somewhat an art, particularly for organic materials. Metallurgists have for some time explored the problem of crystal growth in metal systems. Solid state physicists have, for many years, investigated the problem of crystal growth, structure, and behavior in areas related to solid state electronics. Since the prime candidate materials for spacecraft phase change thermal control devices are currently organic materials (the n-paraffins), the investigation of the melting and solidification of these materials becomes quite important. Therefore, this study concerned an investigation of the crystallization of one n-paraffin, hexadecane.

The goal of this investigation is two-fold:

(1) Develop an improved microphotography apparatus for observing the solidification process,

(2) Perform meaningful studies on the crystallization dynamics of the solidification of the n-paraffins.

In regard to the first goal, one author had the opportunity to spend two summers at NASA/MSFC on a NASA/ASEE Summer Faculty Fellowship Program working with Mr. Tommy Bannister and Miss Barbara Richard (Reference 1) in the microphotography area. Based upon his research using the equipment developed at MSFC, a number of equipment improvements were apparent if a new microphotography apparatus was to be developed. In particular, variable magnification and more flexible sample holders potentially would improve any future microphotography experiments.

In regard to the second goal, a decision was made to study in detail the crystallization of hexadecane. The purpose was to gain quantitative information and understanding on the behavior of one material rather than a brief study of many materials.

2.0 LITERATURE REVIEW

Thomas and Westwater (Reference 2) developed one of the early techniques for microscopic observation of both the melting and solidification process of the n-paraffins. Applying the microphotography techniques that Professor Westwater and his students had for some years used in boiling research, they studied the solid-liquid phase transformations of n-octadecane ($C_{18}H_{38}$), n-hexadecane ($C_{16}H_{34}$), and binary mixtures of these two paraffins.

Bannister and Richard (Reference 1) using a somewhat different experimental approach than Thomas and Westwater, performed a comprehensive microphotography survey study of the solid-liquid phase change process in several members of the paraffin family in addition to water. Motivation for this research was supplied by the need to understand the melting/solidification process of the paraffins when used in phase change thermal control devices. Although their investigation was very comprehensive, results were basically qualitative rather than quantitative. However, a significant contribution from their work was the development of the experimental capability to perform quantitative studies using microphotography techniques.

Following the Bannister and Richard study, other investigations have been carried out in the laboratory of the same authors in attempting to quantify the microphotography observations. A fairly detailed study of the

melting/solidification process of water was conducted (Reference 3), however, it is yet to be published. Golden, Richard and Bannister (Reference 4), conducted a quantitative study of the melting process of hexadecane using the original microphotography equipment of Bannister and Richard, but with an improved test cell.

Although several other microphotography studies of the melting/solidification process of organic materials have been reported in the last several years, they have not been motivated by phase change thermal control and have been primarily qualitative in nature rather than quantitative.

The next section of this report describes the experimental equipment used in the present investigation.

3.0 EXPERIMENTAL EQUIPMENT AND PROCEDURE

As stated earlier, one of the goals of this investigation was to develop an improved microphotography apparatus for the solidification studies. Based upon a review of the apparatus used in Reference 1 and upon consultation with a number of optical suppliers, the apparatus shown in Figure 1 was purchased and assembled. (See Appendix A for all figures.)

The essential components of the microphotography equipment consisted of

- 1) Zeiss Stereo Microscope Basic Body IV & Attachments
- 2) Focusing Eyepiece with Objective
- 3) Bolex H-16 Movie Camera
- 4) EMDECO Camera Drive for the Bolex H-16
- 5) EMDECO Time Lapse Control Package
- 6) Phase Change Test Cell
- 7) Micrometer Rack and Pinion for Positioning the Test Cell
- 8) Light Source and Transformer
- 9) Refrigeration System and Constant Temperature Bath for Supplying Cooling Fluid to the Cell

A detailed equipment list is given in Appendix A for all optical components purchased from outside suppliers.

Figure 2 shows a drawing of the sample test cell used in this investigation. The cell was square and was constructed of plexiglas with copper heat exchangers mounted on both the top and bottom. Two small thermocouples were inserted through the cell walls at different heights such that the thermocouple

junction was centered in the sample material. In addition to the thermocouples inserted in the cell, the temperature of both copper heat exchangers was monitored throughout all runs with thermocouples. Epoxy glue was used to seal the square plexiglas cell walls together. Both epoxy glue and machine screws were used to mount the square cell on the circular plexiglas flanges. A bolt circle of six brass screws was then used to bolt the plexiglas flanges to the copper heat exchangers. Thermal control of the test cell was maintained by use of fluid loops and heat exchangers (a cold fluid for the bottom heat exchanger and an ambient fluid for the top).

The heat exchangers were designed such that one fluid inlet was positioned in the center of the heat exchanger and four symmetric outlets were attached to the edges of each exchanger. If only one outlet was used, it was hypothesized that fluid channeling might produce asymmetric top and bottom temperature profiles and thus introduce errors into the crystalline growth rate measurements. Throughout all the runs, the cooling water to the top heat exchanger was maintained at a constant temperature by passage through a constant temperature bath. The temperature of the cooling water to the bottom heat exchanger during an actual run was maintained at essentially a constant temperature (excluding the early transient period) by a refrigeration unit and a constant temperature bath.

The four cell thermocouples were connected to a four

channel strip chart recorder for monitoring throughout a run.

The hexadecane used in this investigation was practical grade supplied by Eastman Organic Chemicals. Prior to the experimental runs, the sample material was degassed by heating and solidification steps.

The time lapse photography framing rate was 2.14 seconds per frame for all experimental runs. Prior to actually taking experimental data, a series of photographic runs was performed to search for the optimum settings on both the microscope and light source for satisfactory time lapse photographs. The results from this initial study are summarized in Table I. These settings were used in all subsequent experimental work.

Table I

Optical Settings for the Time Lapse Photography Studies

| | |
|------------------------------|--------------|
| camera exposure time | 0.25 seconds |
| f opening of microscope tube | 12 |
| f opening of light source | 7/16 |
| current to light source | 4.5 amps |
| magnification | 20X to 40X |

The experimental procedure for a given run was as follows:

- 1) Load the bolex camera with film (Plus-X black and white), adjust all constant temperature baths for the correct temperature setting, check all optical alignment and start

the strip chart recorder.

2) The fluid loop system was then started and the temperatures of both the top and bottom heat exchangers allowed to stabilize. During this period before the actual run, the fluid to both the top and bottom of the cell was at ambient temperature.

3) After the cell had stabilized thermally, the fluid to the bottom junction was switched by a system of valves and the bottom heat exchanger was then cooled by the new cold fluid. At the same time, the time lapse photographic equipment was started.

4) After the initiation of crystal growth, it was necessary to raise the field of focus in the cell in order to follow the crystals as they grew. This was accomplished by having the cell mounted on a rack and pinion with micrometer settings. The initial cell position was recorded and its new position after being lowered was also noted.

5) The run was allowed to continue until the solidification interface passed the first thermocouple.

6) After the run was completed, the strip chart recorder readings were then converted into temperature versus time plots.

7) Upon completion of step (6), the equipment was then prepared for a new run. Approximately six to eight runs can be made with a 50 foot reel of film. After completion of the reel, the film was then removed from the camera for processing.

The important region in this study was the solid-liquid interface as the interface moved up the cell during the cooling of the bottom heat exchanger. Beautiful crystal fields (crystals of various sizes and shapes) were observed in the region of the interface as the interface progressed. Early runs were made at a magnification such that a multitude of crystals were present in the field of focus. Several later runs were made at a higher magnification in order to follow the growth pattern of individual crystals.

4.0 RESULTS AND DISCUSSION

Analysis of the results of this investigation will be divided into the following topics for presentation and discussion:

- 1) Temperature versus Time Behavior for Each Thermocouple in the Cell
- 2) Average Interface Velocity versus Time
- 3) Average Peak Height Versus Time
- 4) Individual Crystal Growth Rates Throughout the Run
- 5) Qualitative Observations on Crystal Morphology Throughout the Run

A total of eight experimental runs were performed with seven considered quantitatively acceptable. Run 1 was rejected for data reduction purposes due to equipment difficulty; however, this run was included in the qualitative analysis section. The primary experimental parameter that was varied throughout this investigation was the temperature of the cooling water to the bottom test cell heat exchanger, which in turn produced a variation in the paraffin solidification growth rate.

At the completion of the study of hexadecane it was decided to evaluate the differences between the results produced with hexadecane and that of another n-paraffin, octadecane. Therefore, Run 8 was made using octadecane as the test material. Therefore, a significant difference in results will be noted between Runs 2 through 7 (hexadecane) and Run 8 (octadecane) throughout the following discussion.

4.1 Temperature Versus Time Behavior

Figures 3 through 9 (Runs 2 through 8) show the temperature versus time response for each thermocouple attached to the test cell. The seven runs can be broken

down into the following groups depending upon the rate of crystalline growth (determined by the steady state temperature of the bottom thermocouple):

| Growth Rate | Run Number | Steady State Temperature of Bottom Thermocouple |
|--------------|------------|---|
| Slow | 2,3 | 35-38°F |
| Intermediate | 4,5 | 28-30 |
| Fast | 6,7,8 | 22-24 |

A number of observations can be made regarding all seven runs as a group. First, by carefully observing the temperature versus time behavior of thermocouple 2 in the test cell (see Figure 2 for thermocouple designations), it can be seen that the slope of the curve flattens as the interface approaches and crosses the thermocouple. This behavior is reasonable since the heat of solidification must be removed from the liquid before solidification can take place and the crystalline material cooled. Therefore, during the advancement of the interface past the thermocouple, ideally the thermocouple should record a fairly constant temperature. This effect is difficult to see on Runs 2 and 3 (Figures 3 and 4). The flattening effect is obvious on Run 4 (Figure 5). With the high growth rate runs (6,7,8) the effect is masked by the rapid interface growth and all one observes is a slight change in slope of the Thermocouple 2 temperature profile.

An interesting point is the comparison of the reported melting point of hexadecane (approximately 68°F) with that recorded by Thermocouple 2 as the solid interface approaches and crosses the thermocouple position. Consider the data

presented in Table II.

Table II

| Run Number | Temperature Range During Passage of the Solidification Front, °F |
|---------------|--|
| 2 | 63-63 |
| 3 | 63-63 |
| 4 | 66.5-65 |
| 5 | 64.5-63.5 |
| 6 | 65.5-63.5 |
| 7 | 66-65 |
| 8 | 77-75 |

The data for Run 8 is naturally different from the data for Runs 2 through 7 since a different material was used in the test cell (octadecane). However, the data for all runs is low in comparison to the reported melting points for hexadecane (68°F) and octadecane (82°F). The melting points of the n-paraffins will depend upon purity and therefore one can partially explain the differences between the data of Table II and the reported values of the melting points. The possibility also exists that some supercooling was present as the solidification front passed the thermocouple and therefore another potential explanation of the low values in Table I.

4.2 Average Interface Velocity Versus Time

The next parameter of interest is the average interface velocity versus time behavior for each run. Since the solid-liquid interface is very jagged, consisting of peaks and valleys with various crystalline shapes, one has difficulty in defining the interface location on any individual frame of film. However, by assuming a reference interface to be located at the base line of the crystals (where individual crystals are no longer defined on a single frame of the film) one may follow this reference interface throughout a given run. By making interface location measurements at two film frame intervals throughout the film and knowing the actual time that elapsed during the two frame period, a film velocity can be computed. This film velocity can then be converted to an actual velocity in centimeters per second by applying the correct magnification factor for the run. This velocity is then termed an average interface velocity.

Figures 10 through 16 present average interface velocity versus time for Runs 2 through 8. As could be predicted from fundamental heat transfer concepts, the average interface velocity decreases with increasing time. As the interface moves away from the bottom of the cell, the heat transfer resistance increases and therefore the velocity decreases. This behavior was qualitatively observed on all runs, however in Runs 3 and 5 the data were particularly scattered. As would be expected, the cooler the bottom heat exchanger temperature, the higher the average interface velocity.

Velocity values ranged from 2×10^{-4} to 14×10^{-4} cm/sec.

With the particular cell design used in this investigation, it was impossible to observe the onset of solidification (we could not focus on the base of the cell effectively). Therefore all data were taken after the onset of solidification and after approximately the first 60 seconds of growth. If any growth with serious supercooling was present, it would have been observed during the very early part of the run (the portion which we could not observe). However, it is possible that the high average interface velocity values during the early part of each run could be attributed to rapid growth due to supercooling in addition to heat transfer considerations. A modification of the test cell will allow evaluation of this problem by permitting the observation of the onset of solidification.

4.3 Average Peak Height Versus Time

On an individual frame of film, an average peak height quantity was defined as the distance from the average interface (defined in Section 4.2) up to an imaginary horizontal line placed at the tips of the crystals leading in the growth. In other words, this quantity gives an indication of the distance that certain individual crystals lead the majority of other crystals forming the interface. Figures 17 through 23 present the average peak height quantity as a function of run time for Runs 2 through 8.

It is interesting to note that the variation of peak height with time is relatively linear during the early

portion of the run (Figures 17 through 23). However, when the solid interface nears Thermocouple 2, this linear relationship is broken with a sharp reduction in peak height which again increases with run time after the thermocouple is passed.

One possible explanation for the discontinuity of the peak height behavior in the region of the thermocouple is from a heat transfer point of view. Near the thermocouple the high thermal conductivity of the metal wires causes heat to be conducted rapidly from the hotter liquid to the cooler advancing solidification front and therefore inhibits the peaks from advancing ahead of the bulk interface. Thus, the thermocouple acts as a thermal barrier to the formation of large peaks.

By comparing Figures 17 through 23 with the steady-state temperature of the bottom heat exchanger for each run, it can be observed that the maximum peak height measured on each run increases as the steady-state temperature of the bottom heat exchanger decreases. Therefore, it can also be stated that the maximum peak height measured on any run increases with increasing crystal growth rate. Consider Table III.

Table III.

| <u>Growth Rate</u> | <u>Run Number</u> | <u>Maximum Peak Height During Run</u> |
|--------------------|-------------------|---------------------------------------|
| Slow | 2, 3 | 40- 70 x 10 ⁻³ cm. |
| Intermediate | 4, 5 | 70-110 x 10 ⁻³ cm. |
| Fast | 6, 7, 8 | 100-250 x 10 ⁻³ cm. |

4.4 Individual Crystal Growth Rates

Another interesting point to consider is the relationship of individual crystal growth rates in comparison to the growth rate of the bulk of the crystals (average interface velocity). Figures 24 through 30 present a comparison of selected individual crystal growth rates at particular times during a run with the average interface velocity.

Now the interesting point here is the fact that certain individual crystals grow much faster at a particular time than the bulk of the crystals. For instance, examination of Run 2 (Figure 24) at a time of 3 to 4 minutes, one finds an average interfacial velocity of about 6×10^{-4} cm/sec while one individual crystal velocity was calculated to be 15×10^{-4} cm/sec (2.5 times greater). This observation is true for all runs save Run 4 (Figure 26) where the individual crystal velocities selected for evaluation appeared to be of the same order as the average interfacial velocity. Examination of all runs indicates that an individual crystal growth rate may be as much as 2.5 to 3.0 times as great as the average interfacial velocity.

A good explanation for the increased individual crystal growth rate at this point is not clear. One could explain increased growth rates on local subcooling of the liquid in the region of the crystal or upon the ease of growth of certain crystal faces over others. This point is worthy of future investigation.

4.5 Qualitative Observations in Crystal Morphology Throughout the Run

Since it would be difficult and costly to include many photographs of the crystalline interface throughout all the runs, the authors will be glad to loan a copy of the film to all those interested in viewing the experimental work. However, some qualitative observations on each run are in order and are given below.

Run 1 - Slow growth rate

(See Appendix C for definition of terms in the following outline.)

1. Crystals start growing thorn like - slowly becoming fat and more numerous.
2. Some crystals have small, thin, pointed crystals attached to a large thorn like base.
3. As the crystal growth approaches the thermocouple wire, the height of the crystal decreases very sharply, and the interface appears as a lawn.

Run 2 - Slow growth rate

1. During initial growth, the interface appears as a group of closely packed shrubs.
2. The slow growth produces an interface that appears like a lawn.
3. Length of the crystal never becomes as prominent as in Run 1, but the growth is very smooth in comparison to Run 1.
4. After the interface passes the thermocouple, the growth becomes very irregular and rough. Crystals appear as thorns with pointed tops.

Run 3 - Slow growth rate

1. Initial interface is fairly smooth.
2. Growth rate increases rapidly, interface appears as mountains and thorns.
3. As growth progresses, the appearance turns bush like.
4. Later in growth, very tall thorns appear.
5. During the latter part of the run, the thorns become much wider.

Run 4 - Intermediate growth rate

1. Early interface appearance like a lawn.
2. Blade shaped crystals become elongated.
3. As growth progresses, the crystals become thorn like and closely packed.
4. Further growth results in a bush like appearance.
5. As the interface approaches the thermocouple, the surface becomes smooth, and then takes on a lawn shaped appearance.
6. After passing the thermocouple, the crystals take on the appearance of tall blades of grass.

Run 5 - Intermediate growth rate

1. Early interface appearance as thick blades of grass.
2. Slowly mountain shapes begin to appear.
3. Thorns begin to grow slowly from the mountains and then the thorns branch out like thistles.
4. Thistles become extremely tall.
5. The interface becomes smooth upon passing the thermocouple.
6. After passing the thermocouple, thorns begin to appear and later become very thick.

Run 6 - Fast growth rate

1. Early interface appears as a lawn.-
2. Thorns begin to appear and become thick.
3. Thorns do not become very tall, but do expand in width.
4. Thorns become very short as the thermocouple is approached.
5. Interface is again grass like after passing the thermocouple.

Run 7 - Fast growth rate

1. Early appearance - fat thorns and some crystals with flat tops, closely packed.
2. Appearance then takes on a grass like appearance with thorns extending from the grass.
3. Thorns become fatter and larger.
4. Same behavior as Run 6 as the thorns pass the thermocouple.

Run 8 - Fast growth rate

1. Early appearance grass like.
2. Very thick and slow and apparent slow growth.
3. Significant growth difference over previous runs, cloud like appearance over mountains, some flat areas.
4. Thorns appear after passing the first thermocouple.
5. Interface takes on a uniform thorn like appearance.
6. Thorns fatten as growth progresses.

4.6 Errors in Photographic Results

All measurements taken from the time lapse film are subject to individual interpretation and therefore some error will be introduced. Measurements such as average interfacial velocity, peak height and individual crystal velocities

depend upon the judgment and definition of the observer and therefore will vary as the people making the measurements change. By this reasoning, the authors feel that an error of 10 to 15% is possible in the above mentioned quantities. In an attempt to assess this error, a number of runs were reduced by two different people using the same definition of interfacial velocity, peak height and individual crystal velocities. Agreement was obtained within the 10-15% margin.

4.7 Comparison with Octadecane

In reviewing the total experimental data, a summary of the differences noted between hexadecane (Runs 2 through 7) and octadecane (Run 8) is appropriate. First, as would be expected, the temperature of thermocouple 2 as the interface approached was significantly different. Secondly, the peak height versus time measured for Run 8 is larger than that for Runs 6 and 7, particularly at later times in the run. Thirdly, the qualitative observations are different for Run 8. In view of the differences noted between the two materials, a detailed study of octadecane is recommended. Further, detailed microphotography studies of other members of the n-paraffin series would be very interesting.

CONCLUSIONS AND RECOMMENDATIONS

The following conclusions are presented based upon the results of this investigation:

- (1) The development of an improved microphotography apparatus (variable magnification and more flexible sample holder) was successful.
- (2) The slope of the temperature versus time curve flattens as the interface approaches the thermocouple.
- (3) The average interface velocity decreases with increasing run time or as the interface moves away from the heat sink.
- (4) The variation of peak height with run time is linear up to the region near the first thermocouple. Near the region of the thermocouple, peak height is reduced and then increases as the interface moves away from the thermocouple.
- (5) The maximum peak height between various runs increases as the crystal growth rate increases.
- (6) Individual crystal growth rates may be as much as 2.5 to 3.0 times the average interface velocity at a given time during a run.
- (7) The solid interface takes on a variety of geometrical shapes depending upon a number of conditions. In particular, heat transfer consideration (interface near a high thermal conductivity thermocouple) perturbs the interface geometry. Thorn like crystals are only observed after the initial growth period.

Based upon the results of this investigation, the following recommendations are presented:

- (1) Future experimental work in the phase-change microphotography area should be centered around
 - (a) quantitative investigation of other members of the n-paraffin family,
 - (b) further definition of the relationship of crystal morphology and experimental conditions,

- (c) rigorous definition and evaluation of any microphotography experiments proposed for space flight (detailed scientific evaluation on earth for comparison with flight results).
 - (d) studies, with high magnification factors, for individual crystal observation.
- (2) Attempts should be made to mathematically model the interface behavior as a function of experimental conditions. To be more specific, it may be possible to predict the crystal type and shape based upon the material and the thermal conditions by using modeling techniques.
- (3) The results of this and future investigations should be evaluated in the light of the best available solidification theory.

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APPENDIX A

Figures

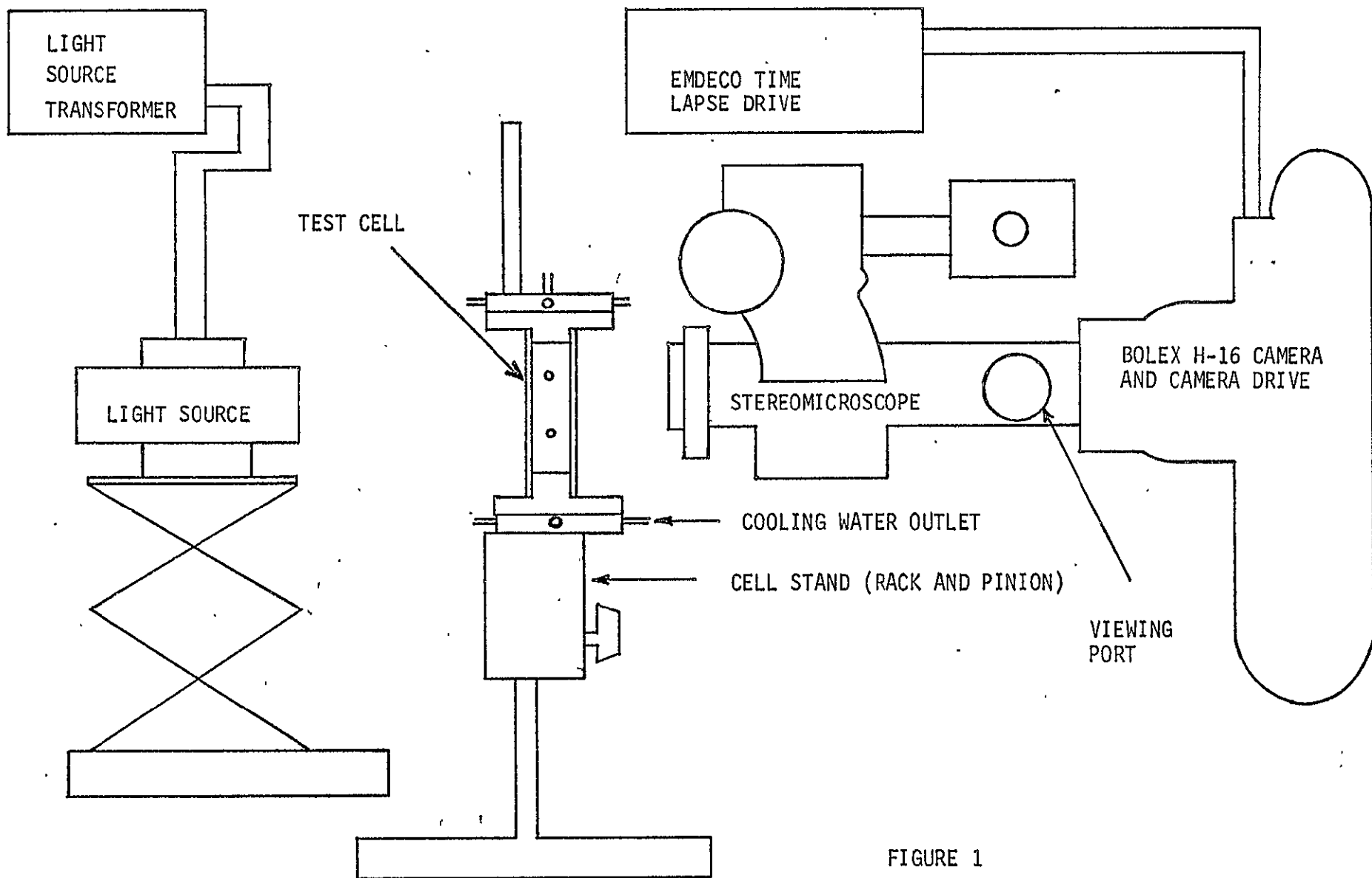


FIGURE 1
TIME LAPSE MICROPHOTOGRAPHY EQUIPMENT
DIAGRAM

FIGURE 2

PHASE CHANGE TEST CELL

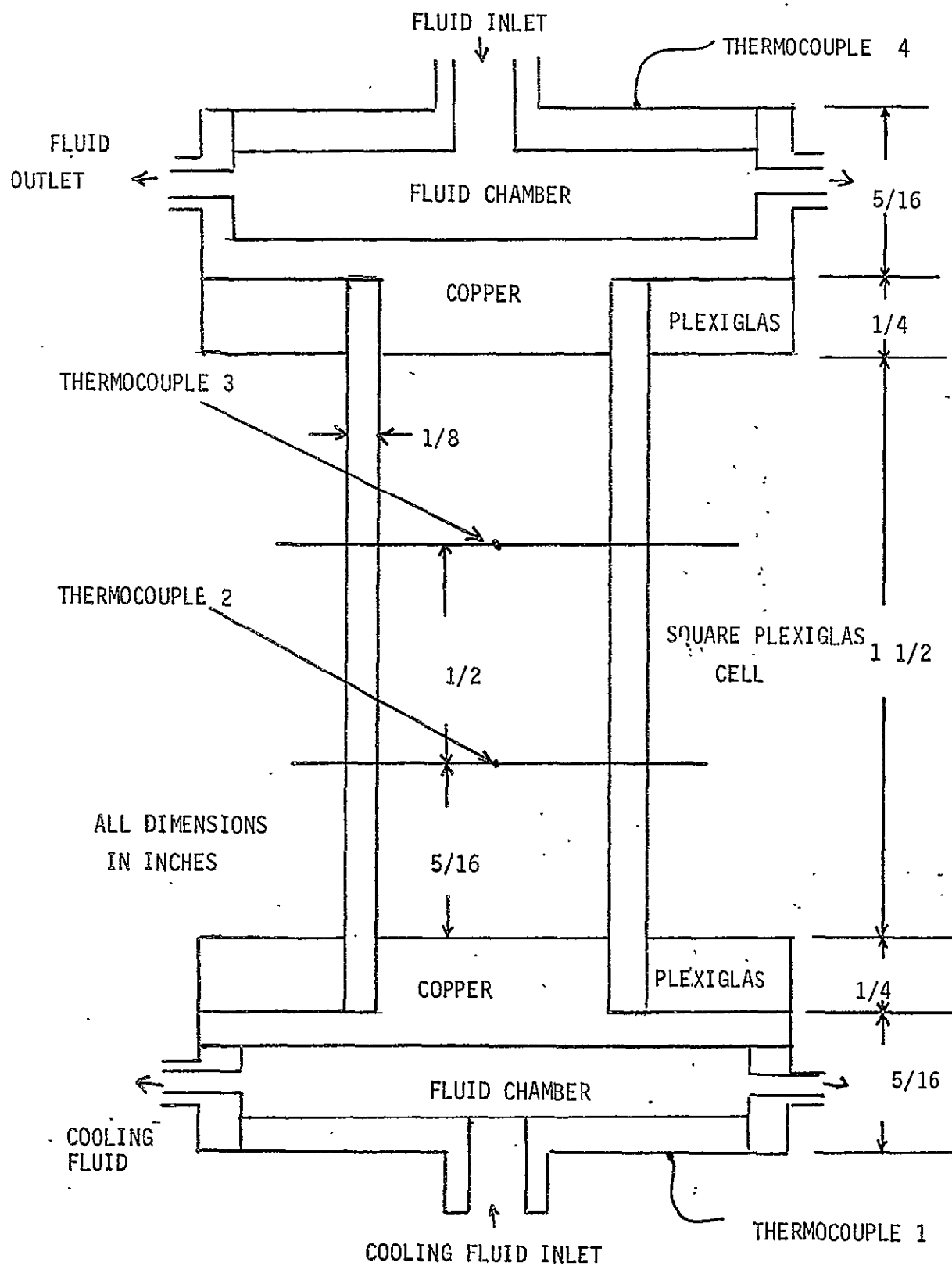


FIGURE 3
TEMPERATURE VERSUS RUN TIME
RUN 2

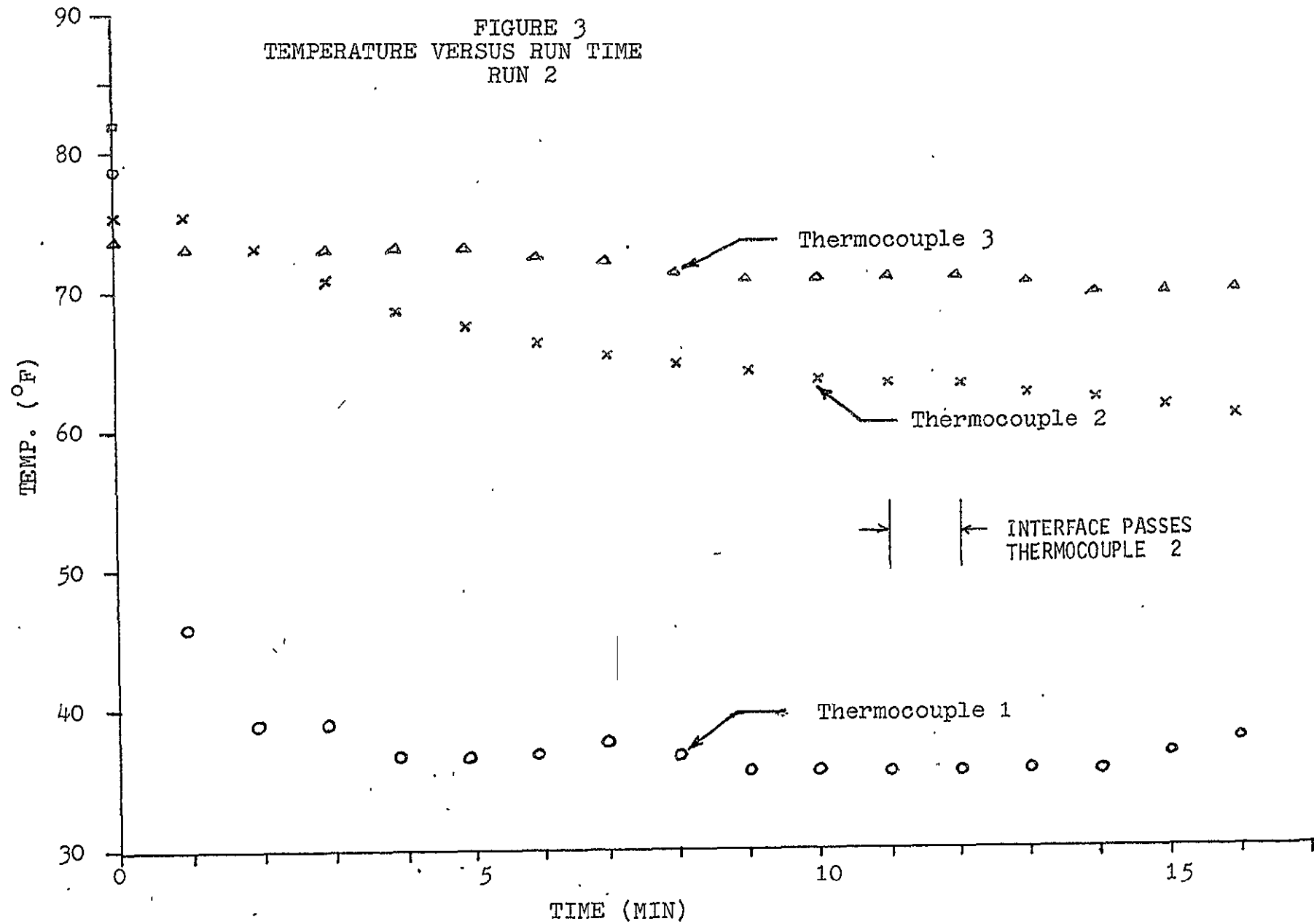


FIGURE 4
TEMPERATURE VERSUS RUN TIME
RUN 3

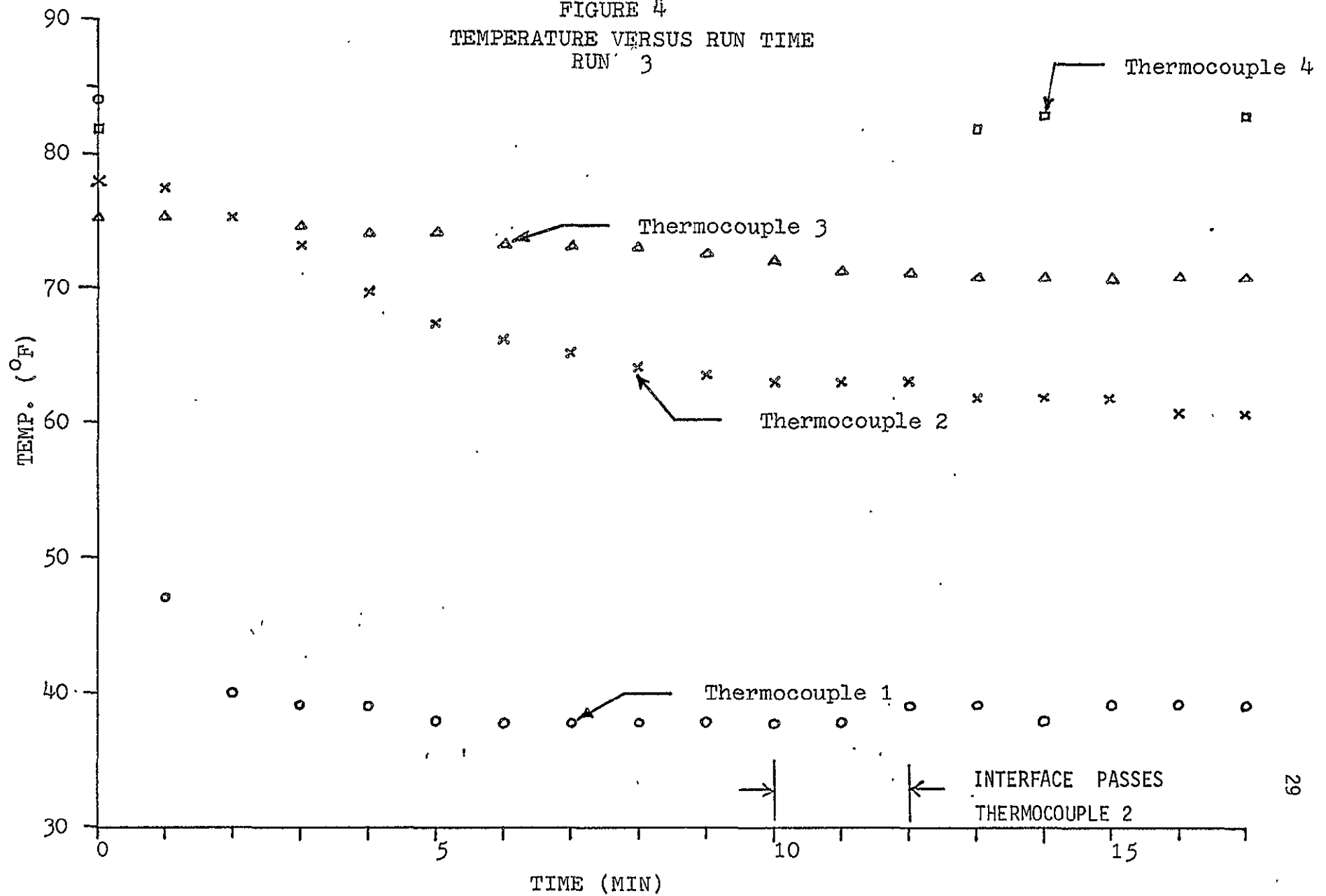


FIGURE 5
TEMPERATURE VERSUS RUN TIME
RUN 4

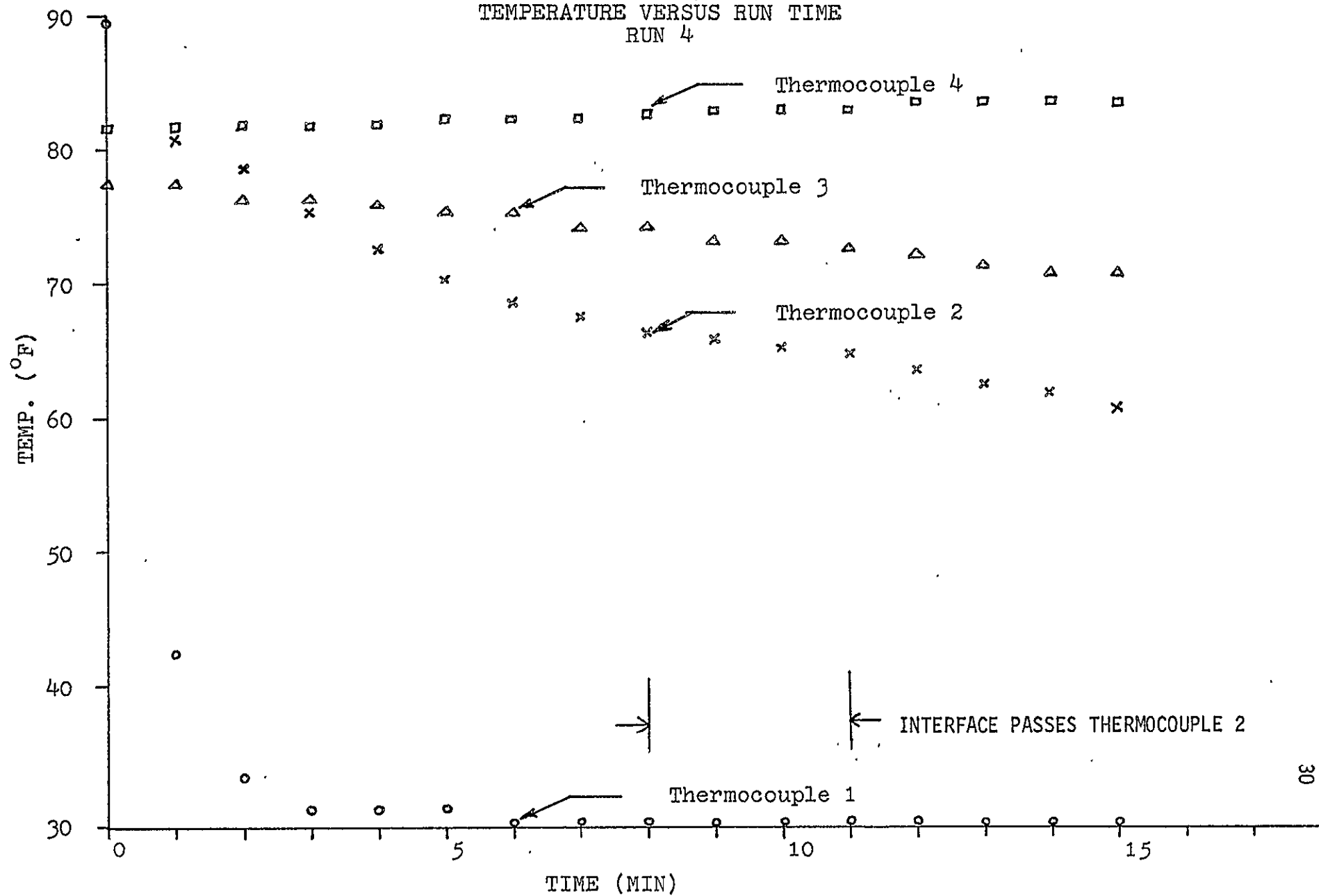


FIGURE 6
TEMPERATURE VERSUS RUN TIME
RUN5

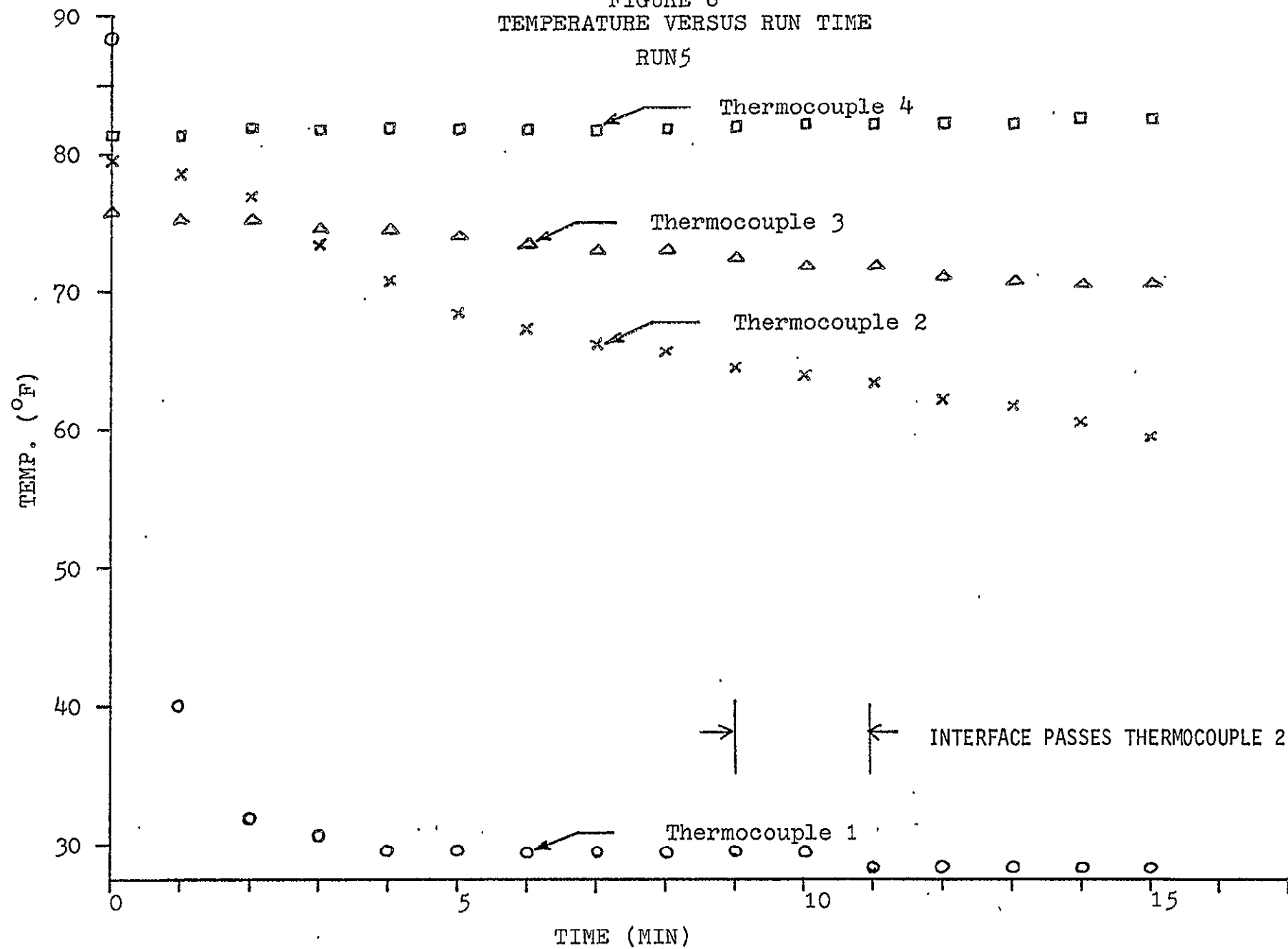


FIGURE 7
TEMPERATURE VERSUS RUN TIME
RUN 6

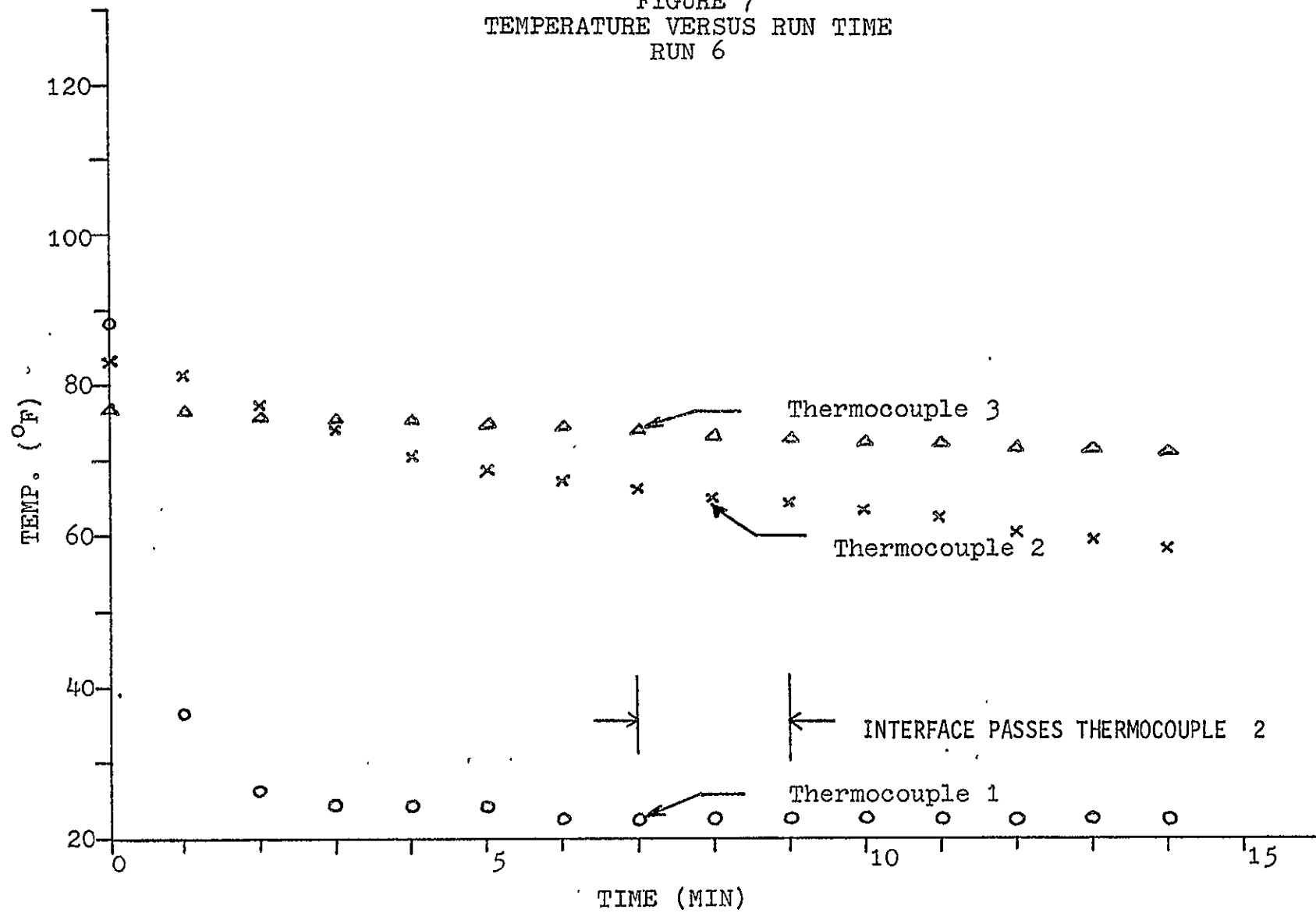


FIGURE 8
TEMPERATURE VERSUS RUN TIME
RUN 7

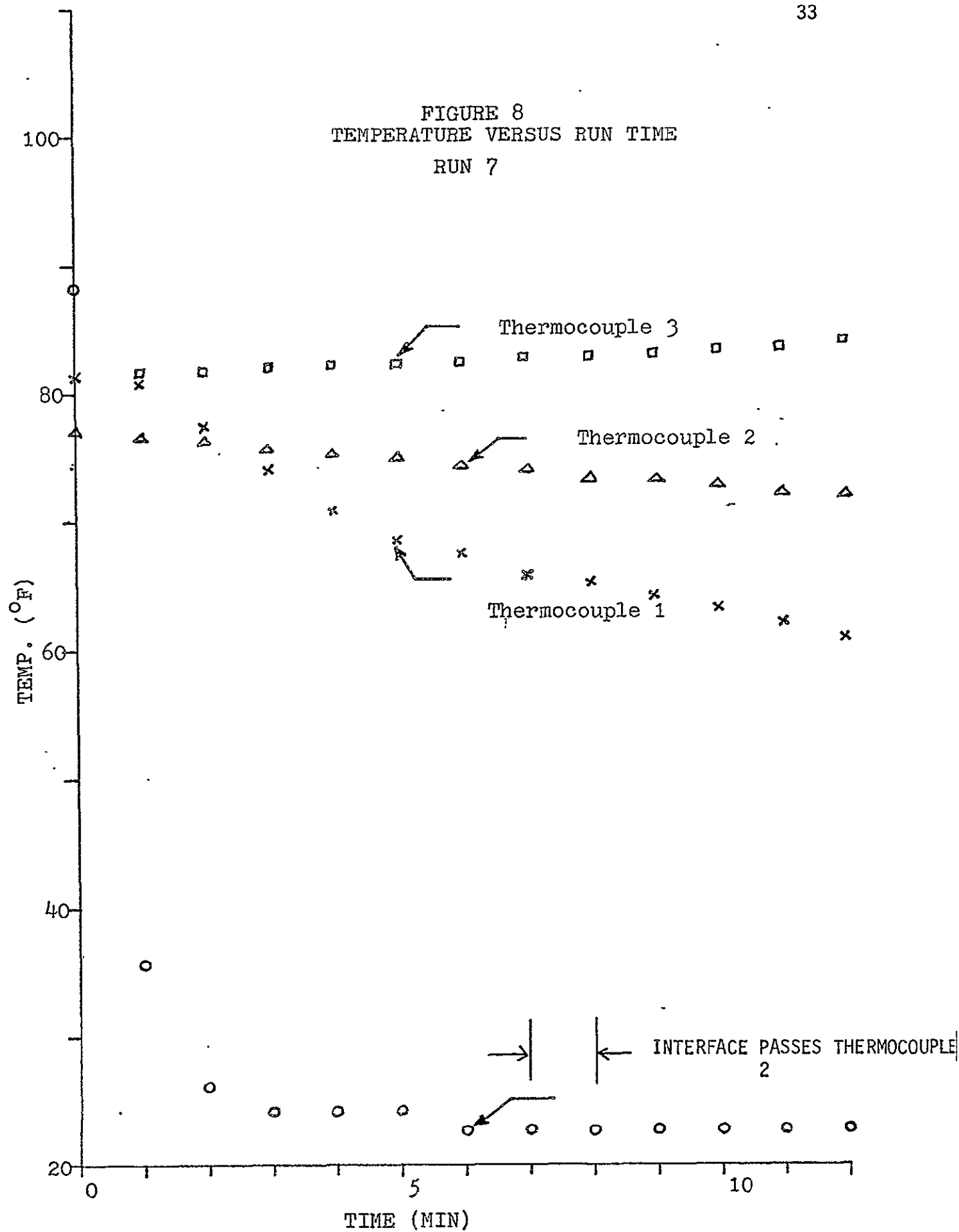


FIGURE 9
TEMPERATURE VERSUS RUN TIME
RUN 8

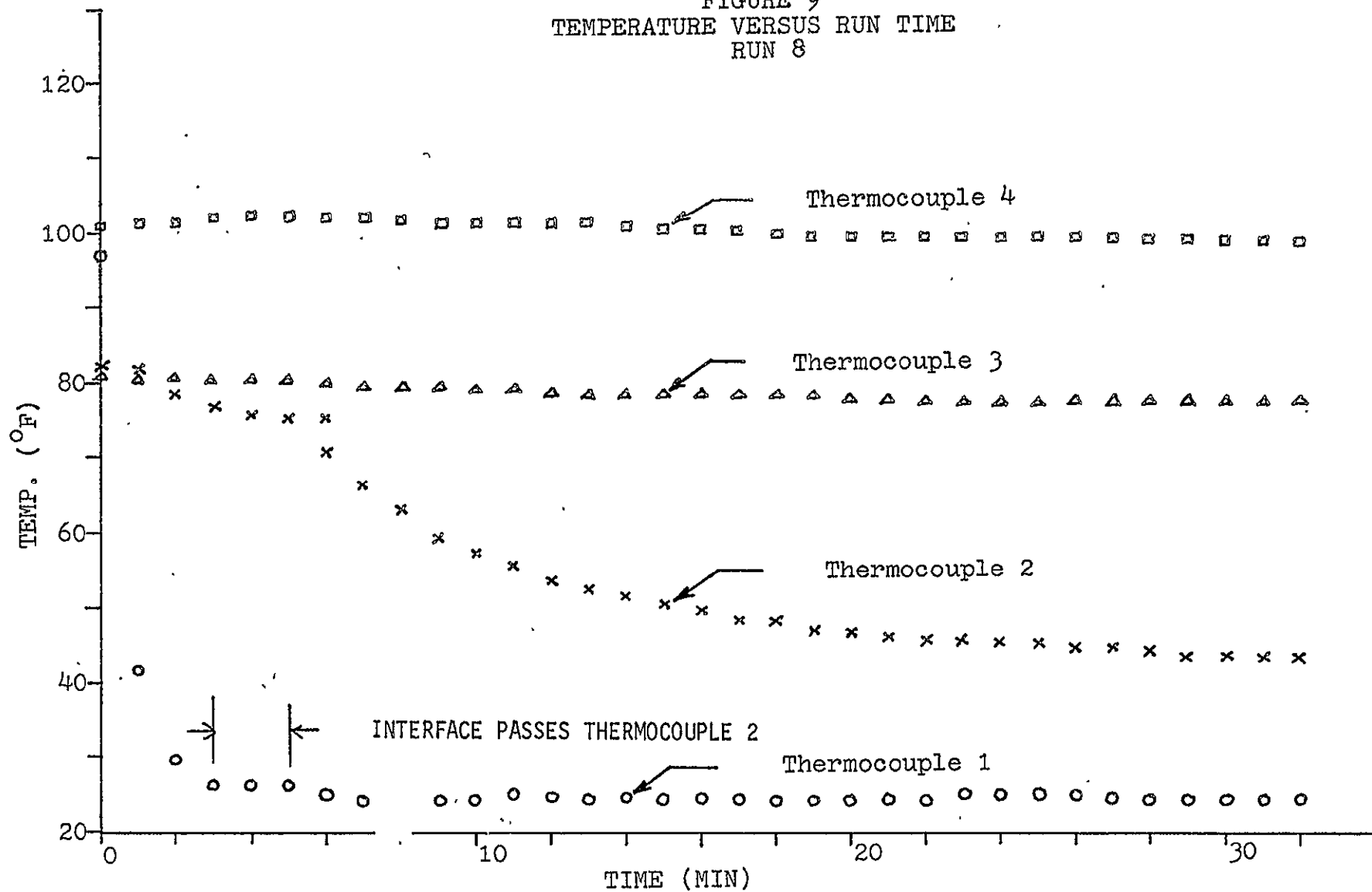


FIGURE 10
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 2

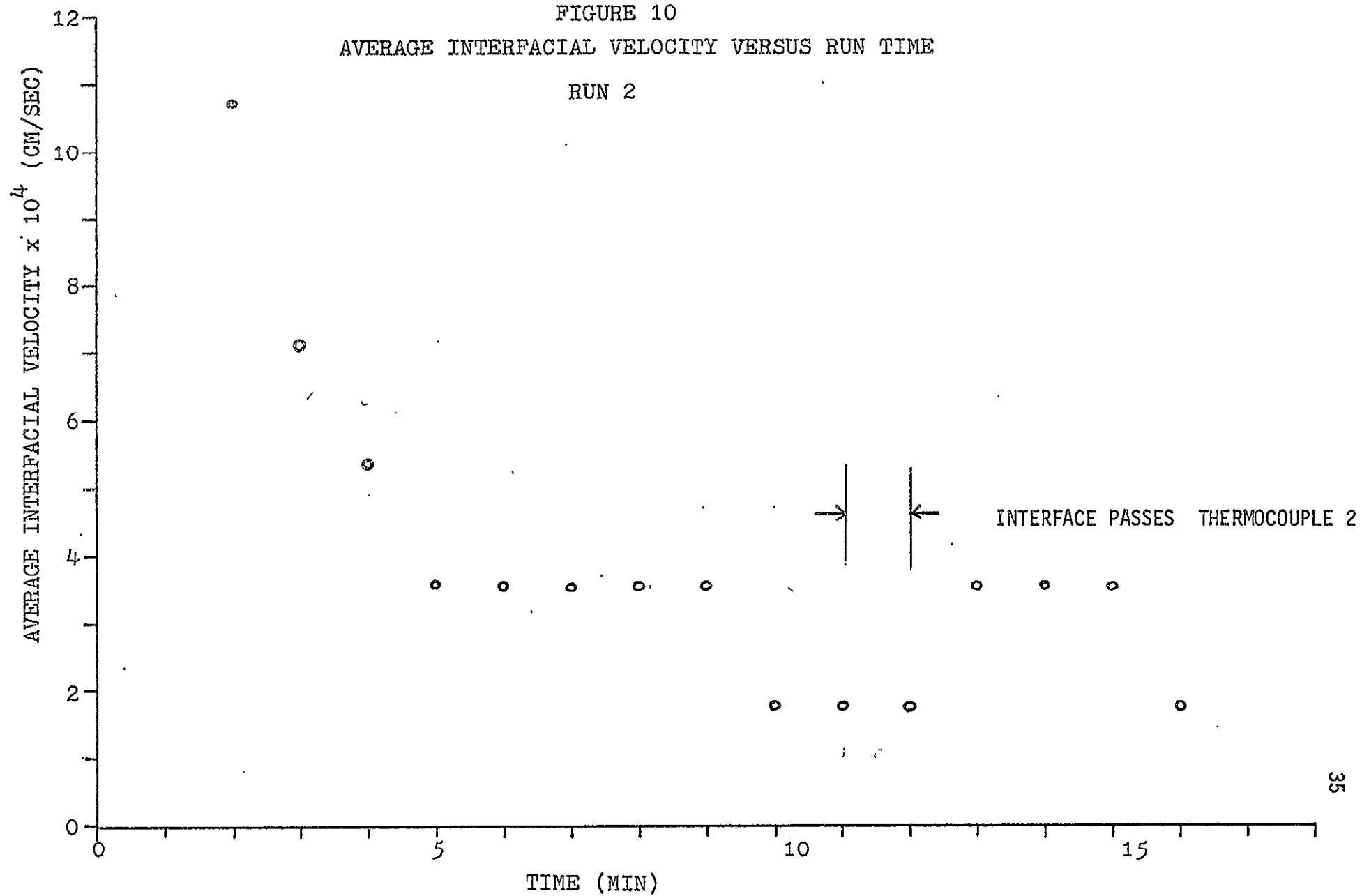


FIGURE 11
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 3

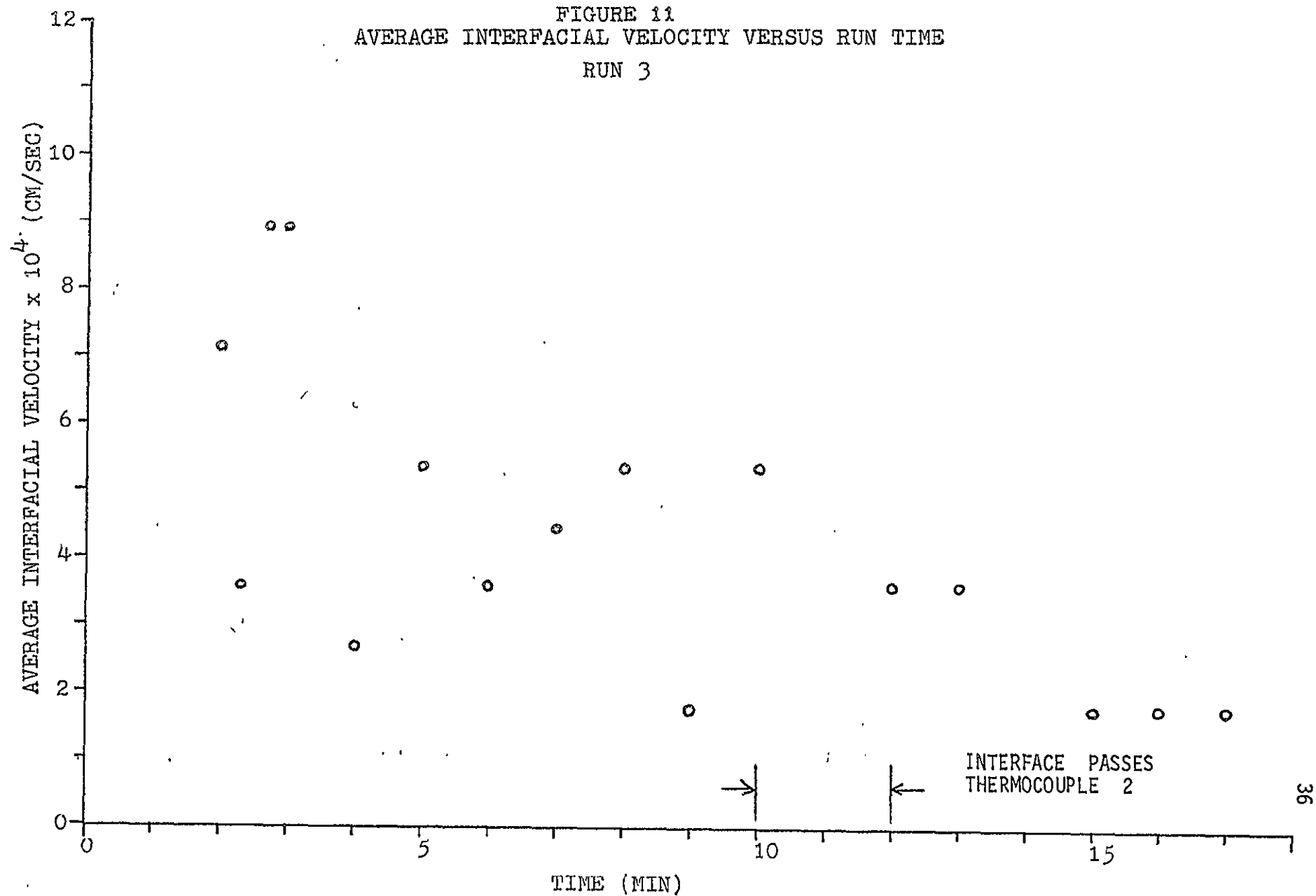


FIGURE 12
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 4

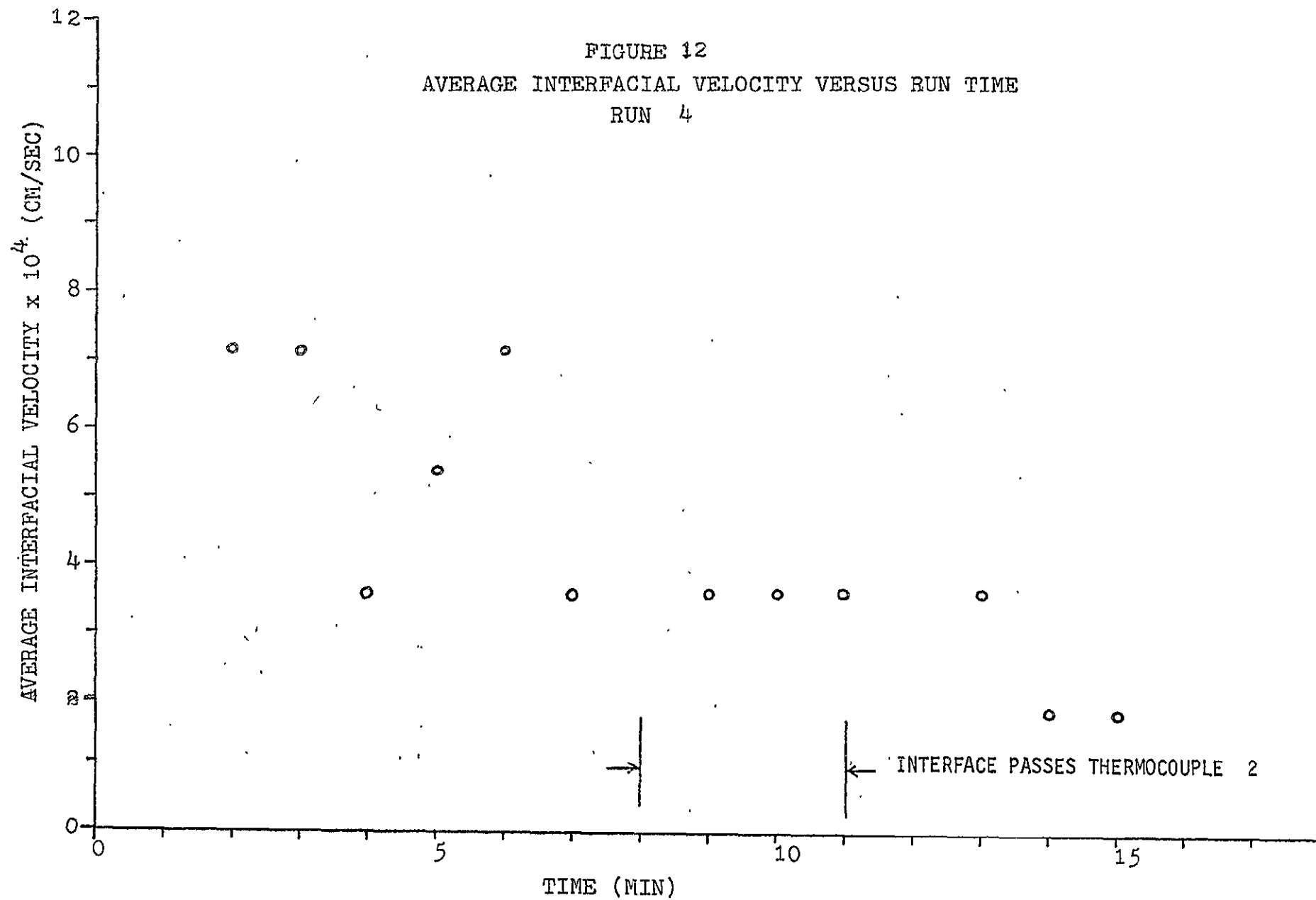


FIGURE 13
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME

RUN 5

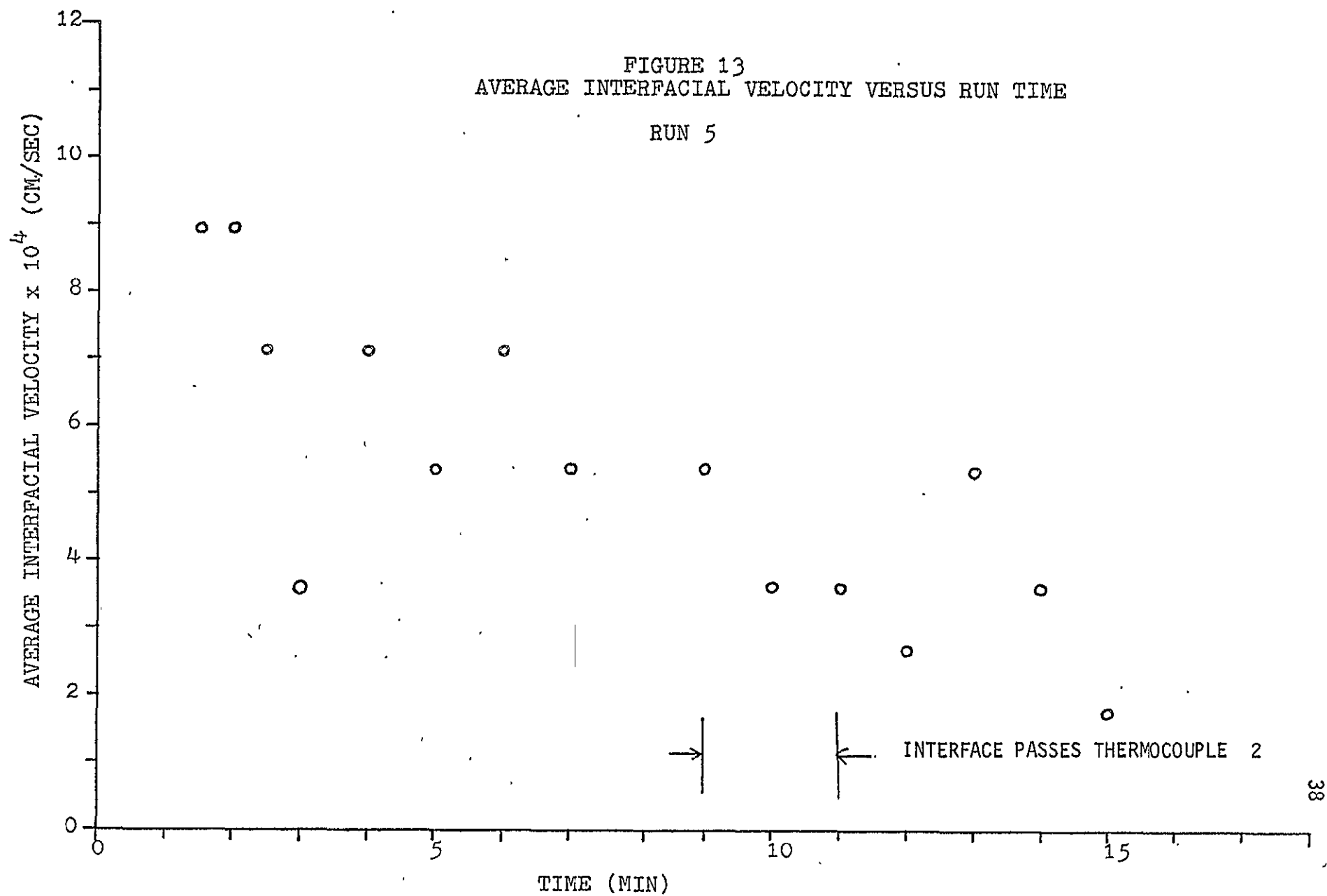


FIGURE 14
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 6

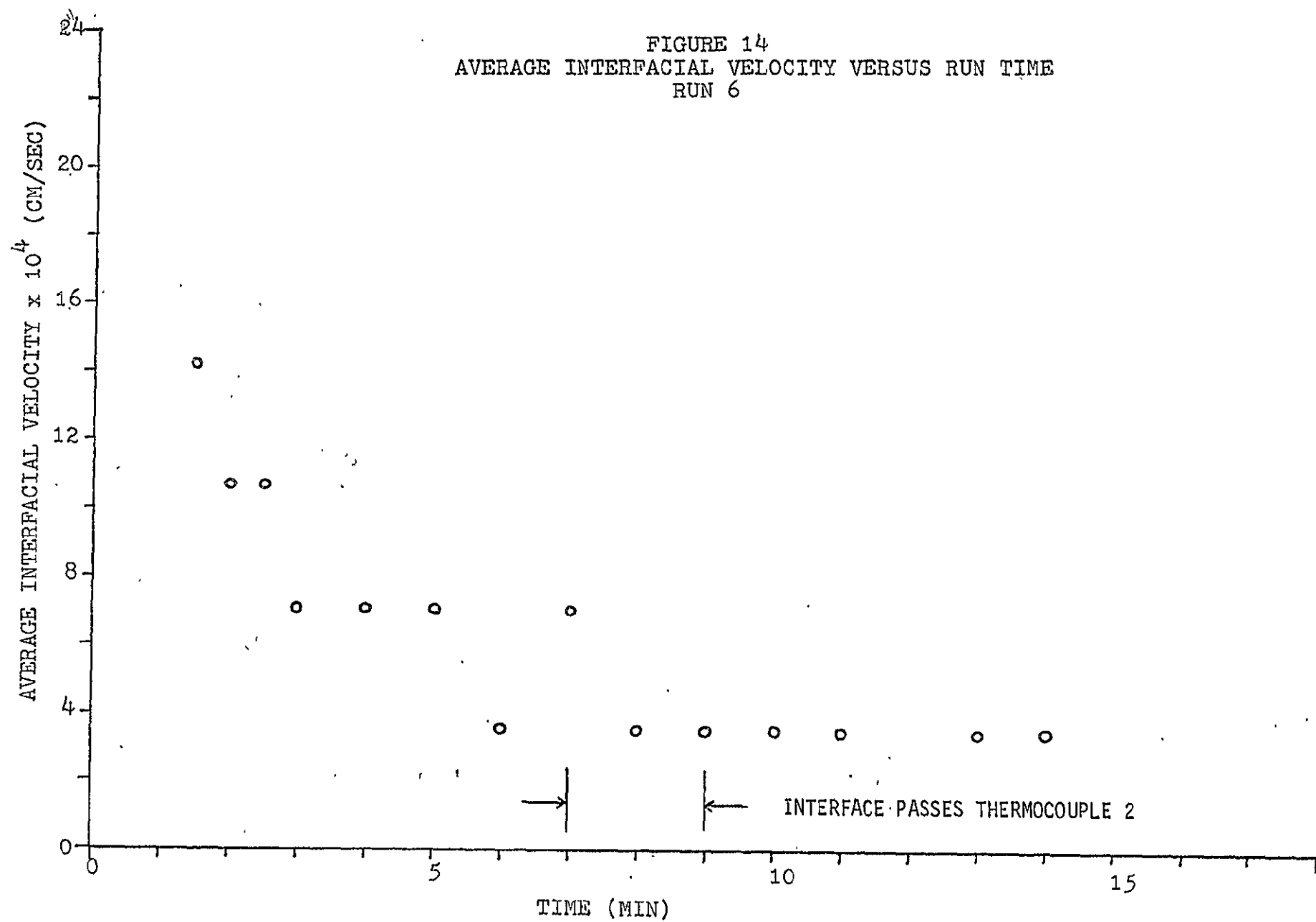


FIGURE 15
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 7

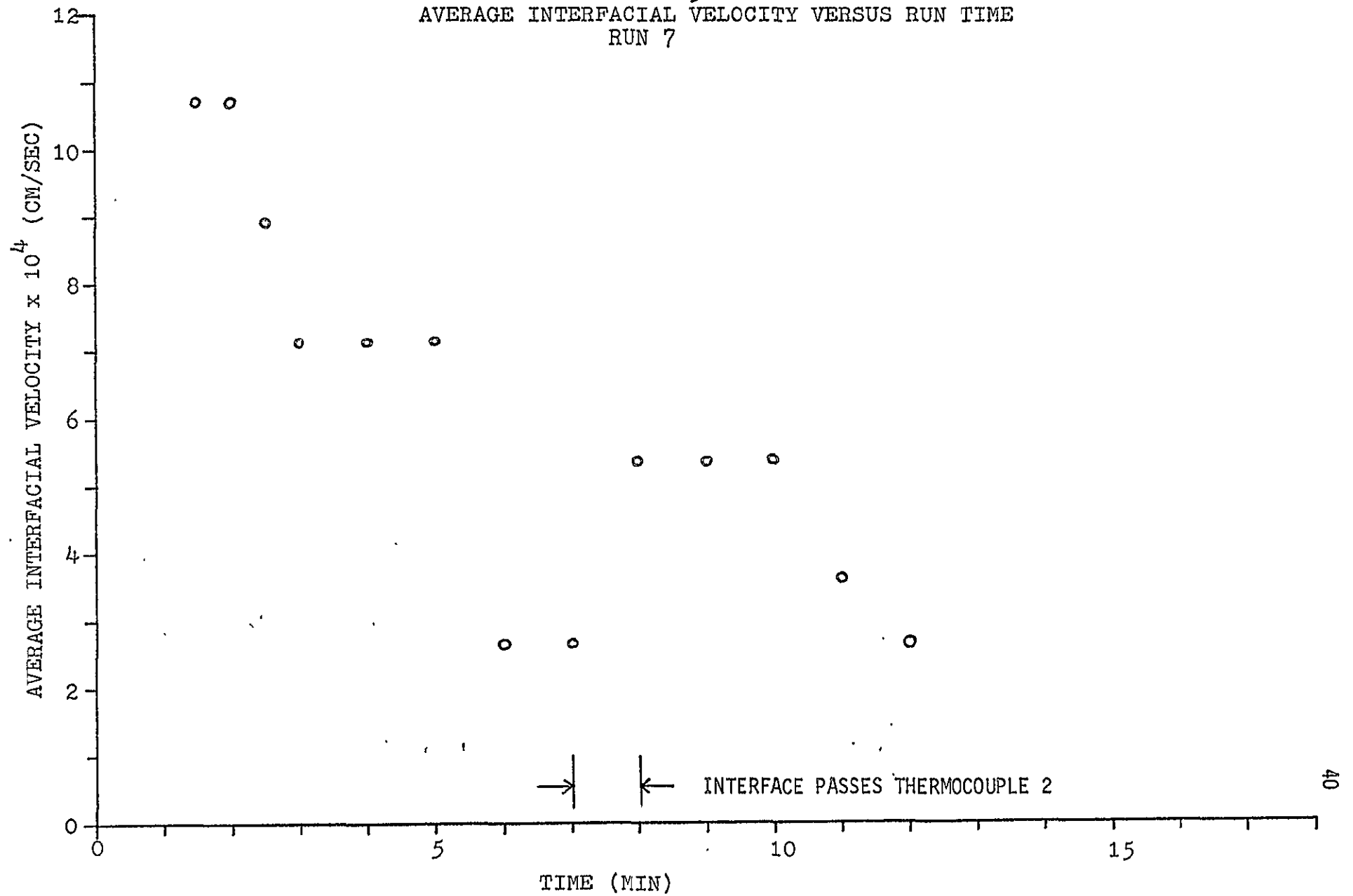


FIGURE 16
AVERAGE INTERFACIAL VELOCITY VERSUS RUN TIME
RUN 8

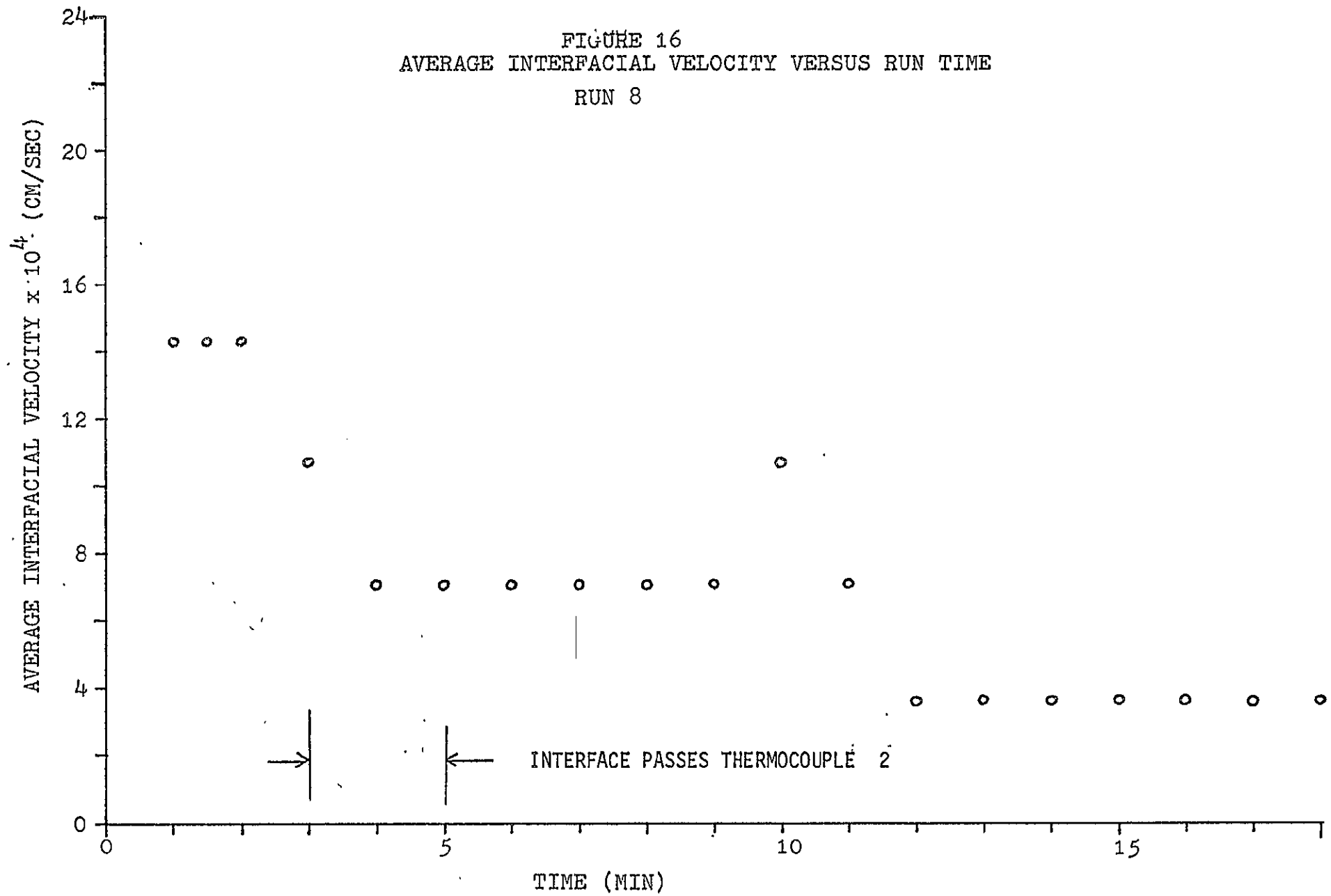


FIGURE 17
PEAK HEIGHT VERSUS RUN TIME
RUN 2

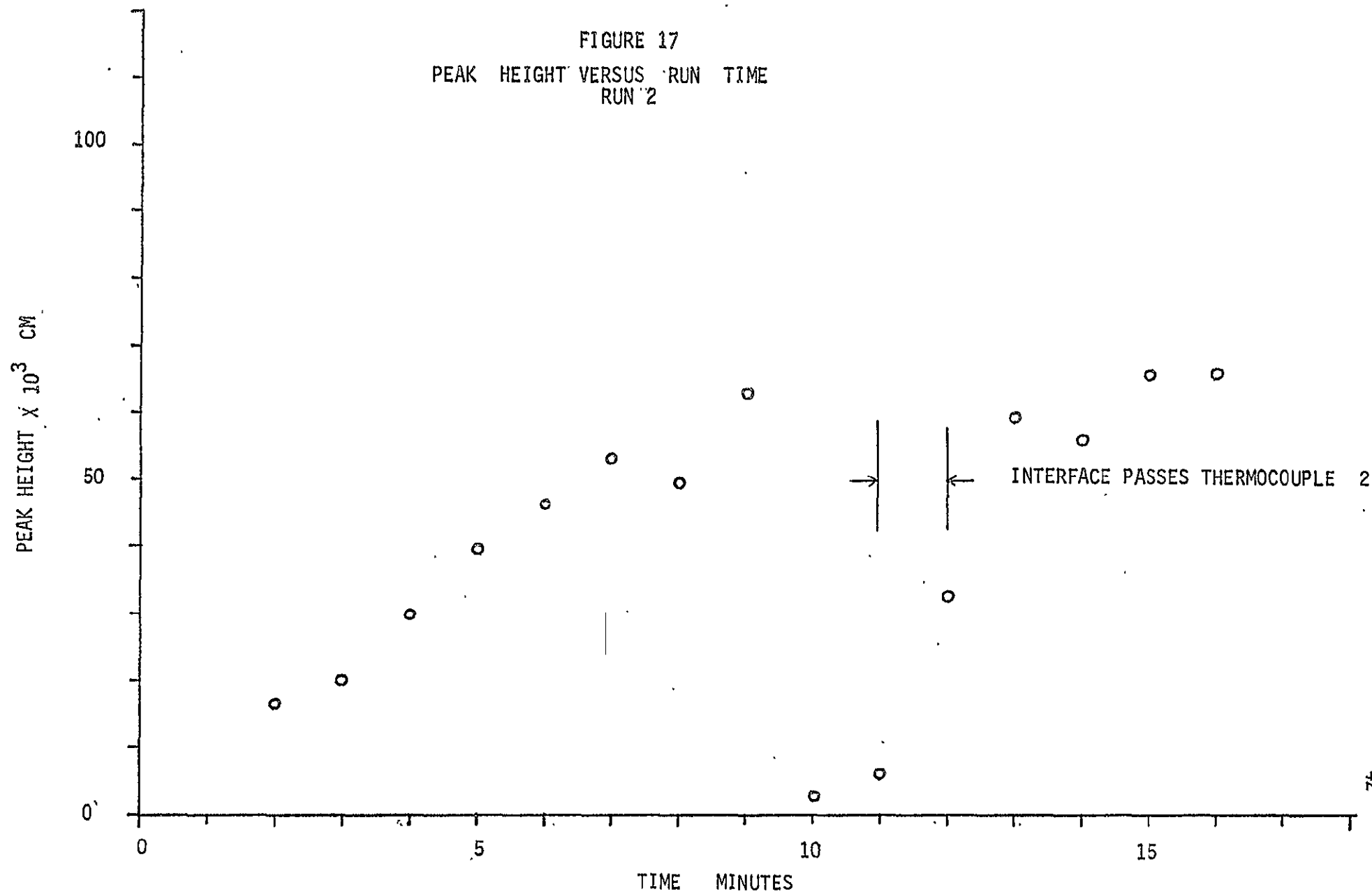


FIGURE 18
RUN 3
PEAK HEIGHT VERSUS RUN TIME

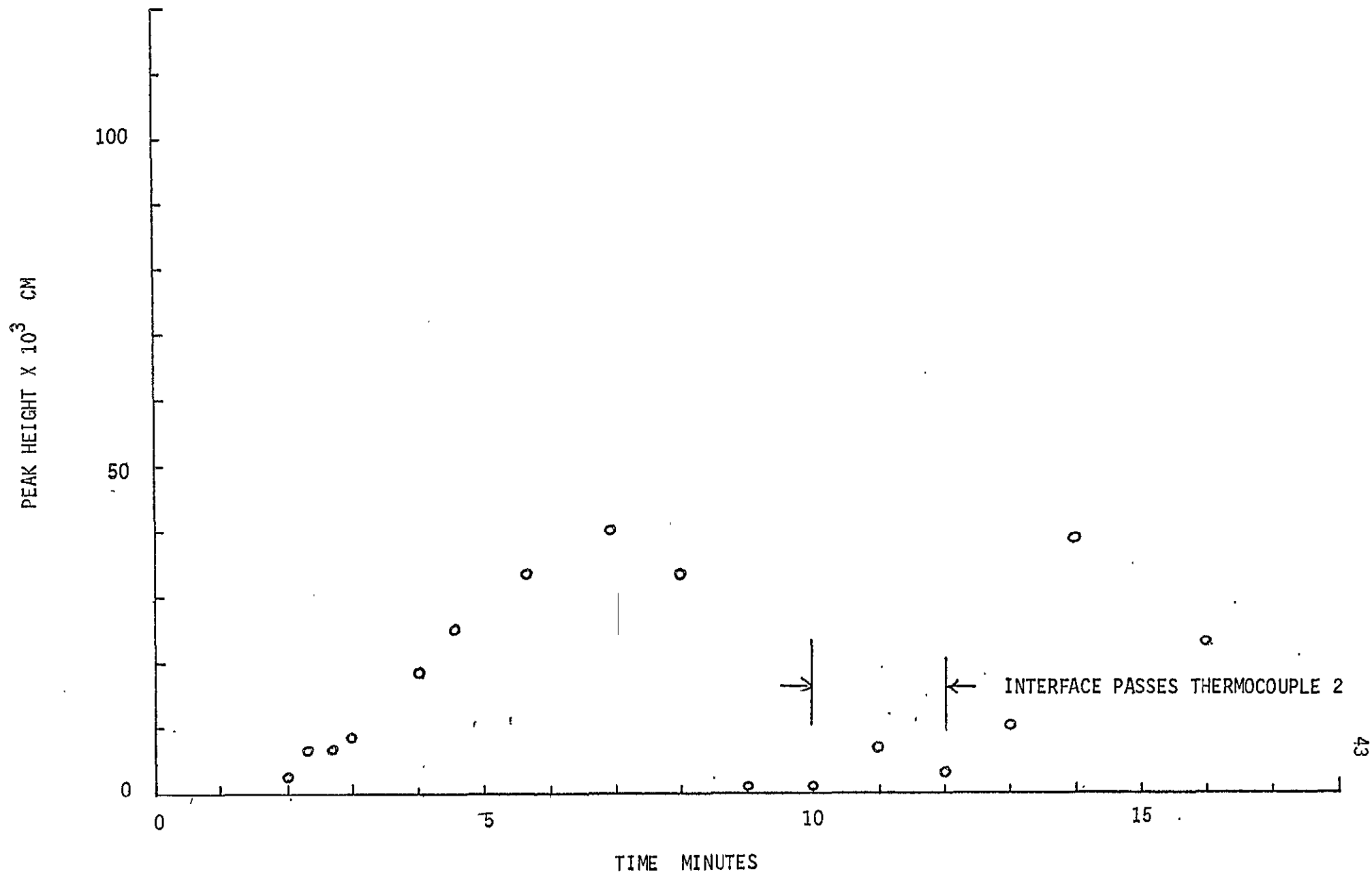


FIGURE 19
RUN 4
PEAK HEIGHT VERSUS RUN TIME

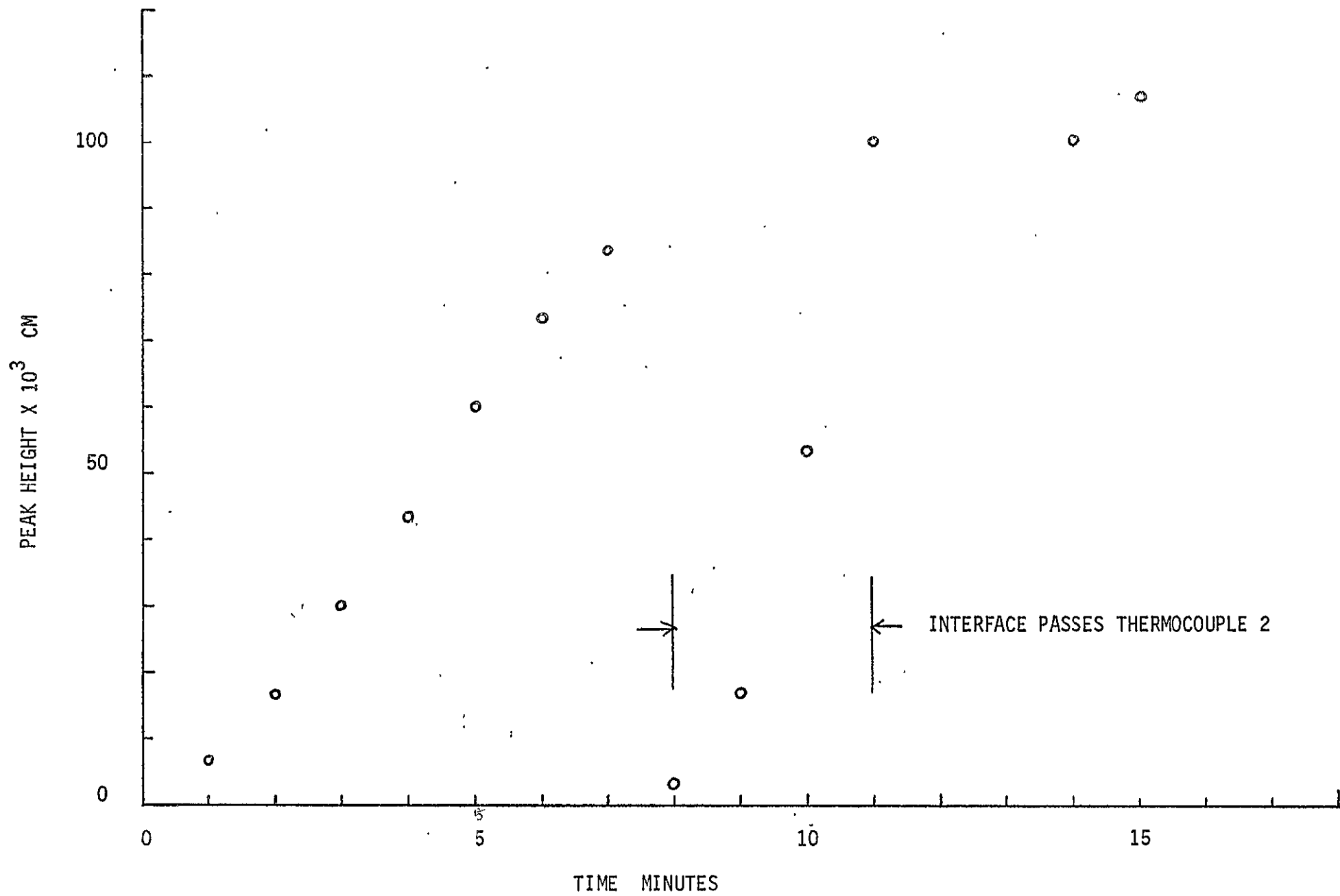


FIGURE 20
RUN 5
PEAK HEIGHT VERSUS RUN TIME

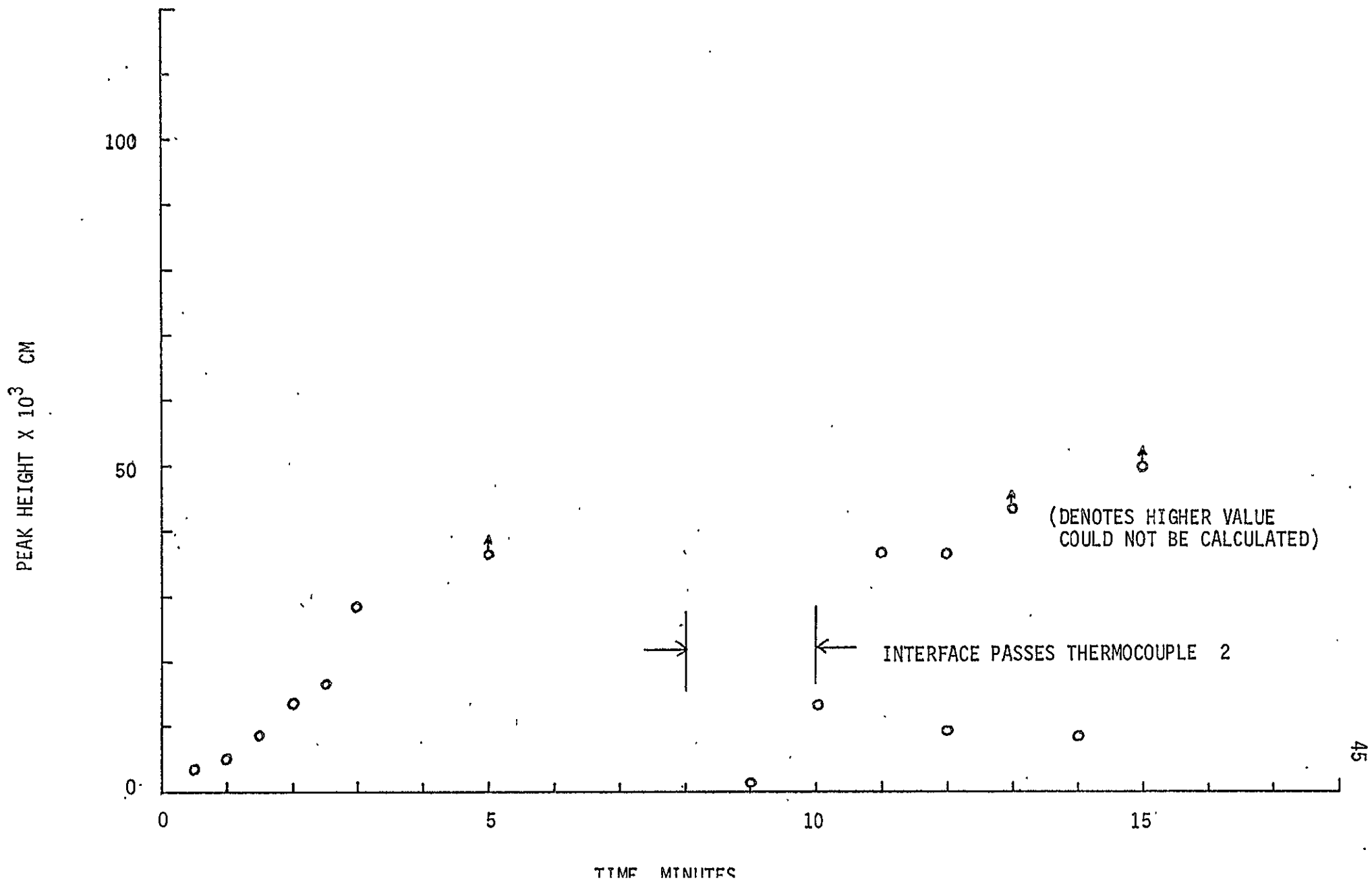


FIGURE 21
RUN 6
PEAK HEIGHT VERSUS TIME

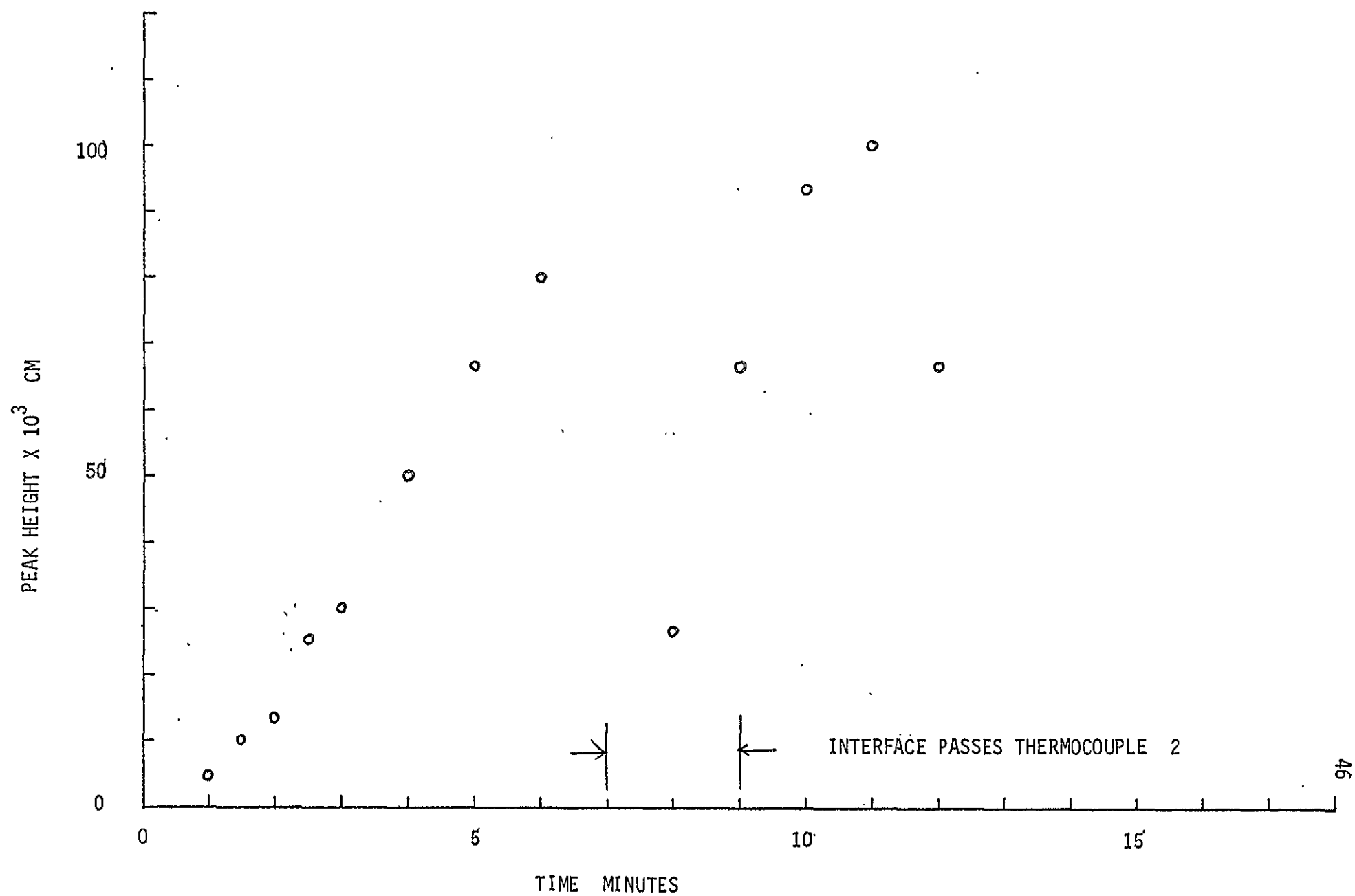


FIGURE 22
RUN 7
PEAK HEIGHT VERSUS TIME

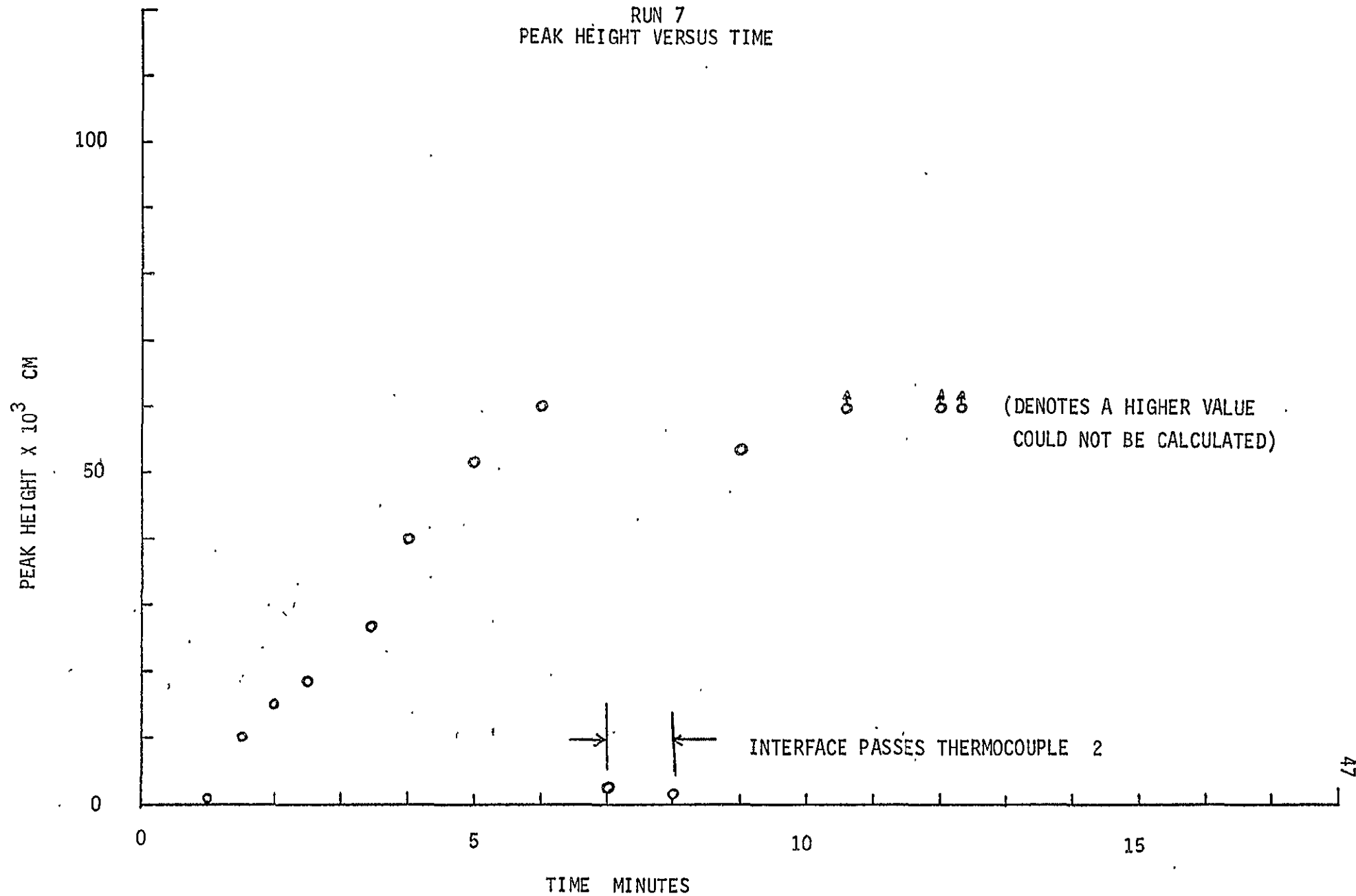


FIGURE 23
RUN 8
PEAK HEIGHT VERSUS TIME

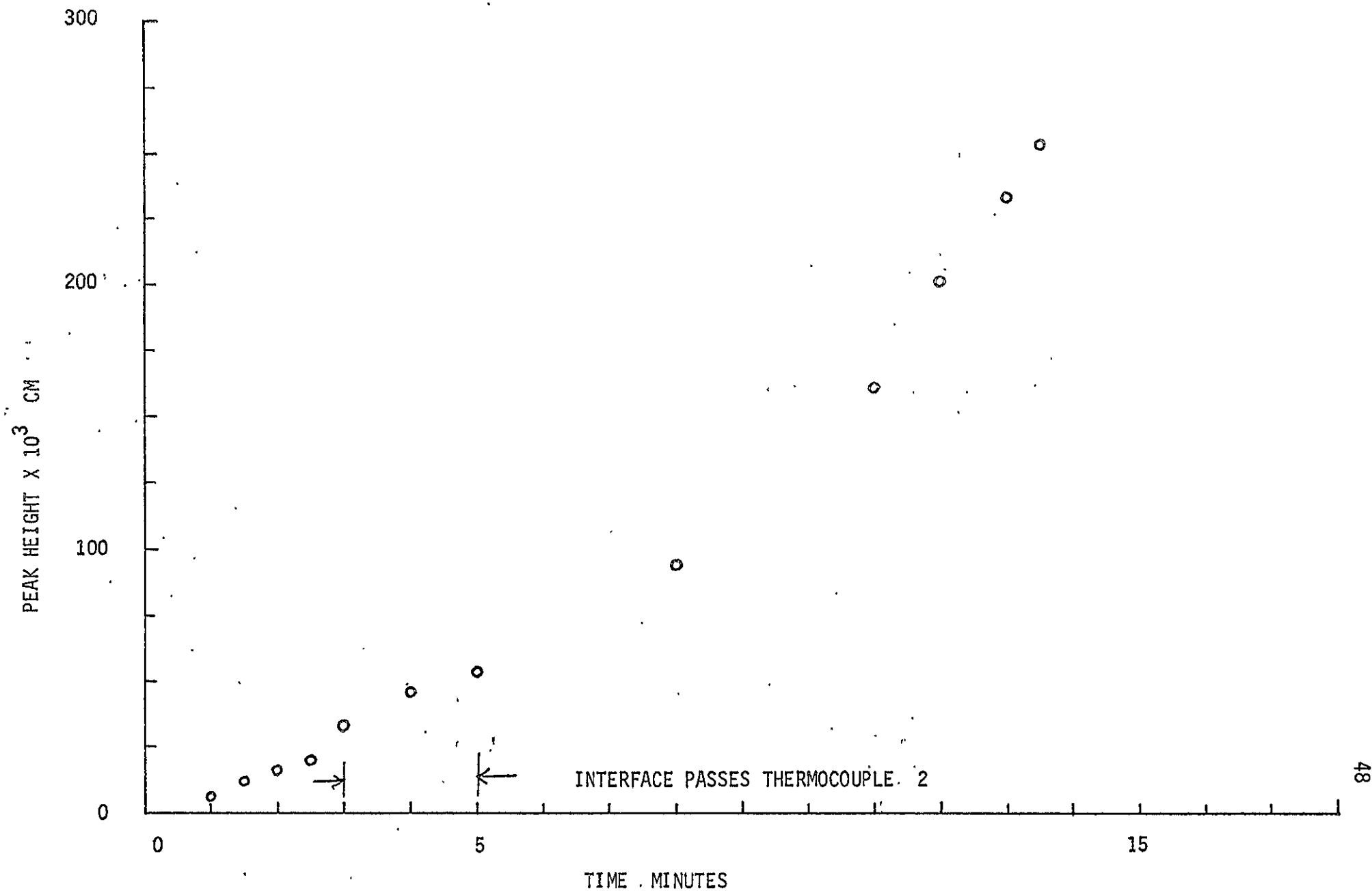


FIGURE 24

RUN 2

AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

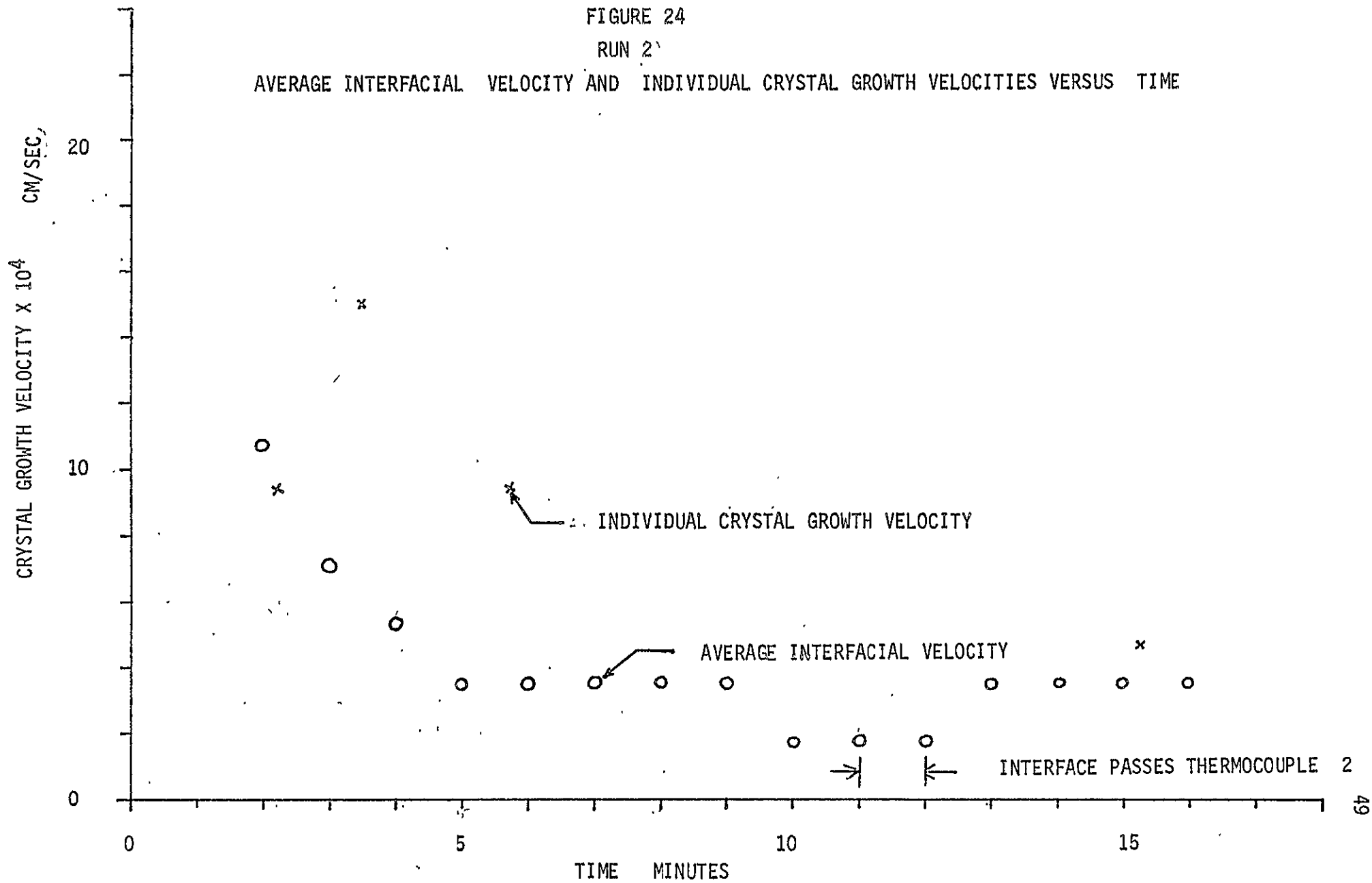


FIGURE 25
 RUN 3
 AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

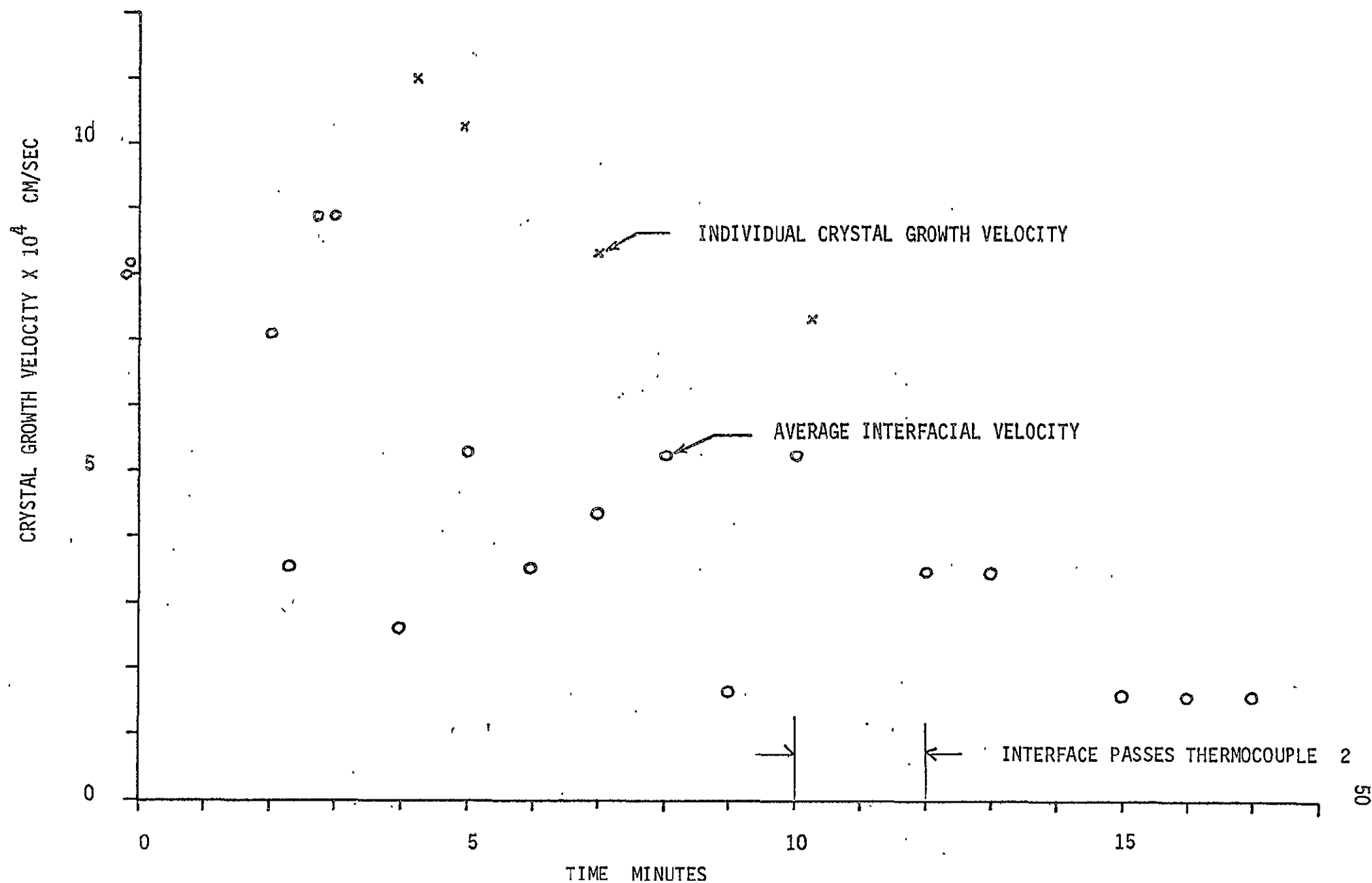


FIGURE 26
RUN 4
AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

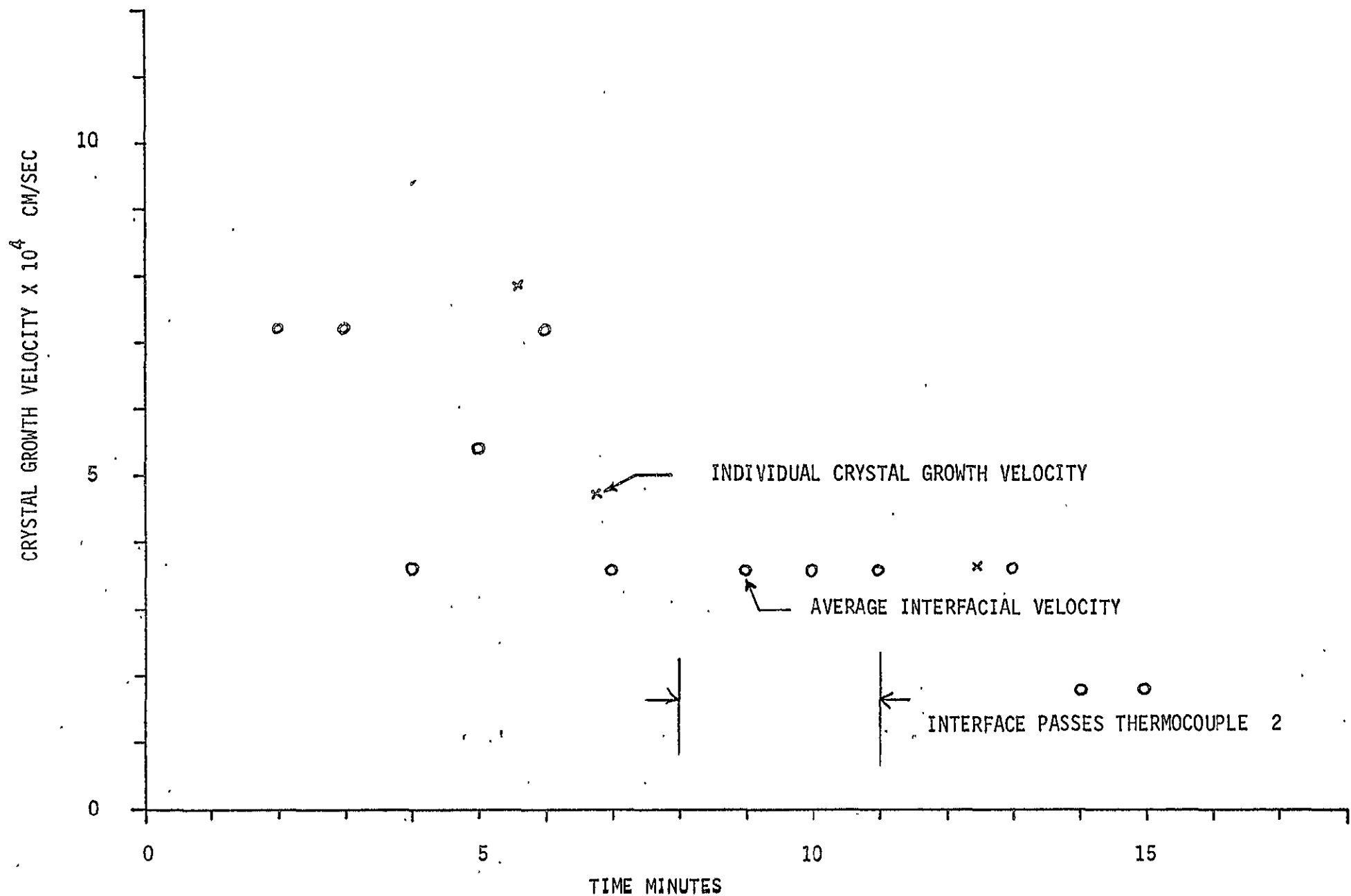


FIGURE 27
RUN 5
AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

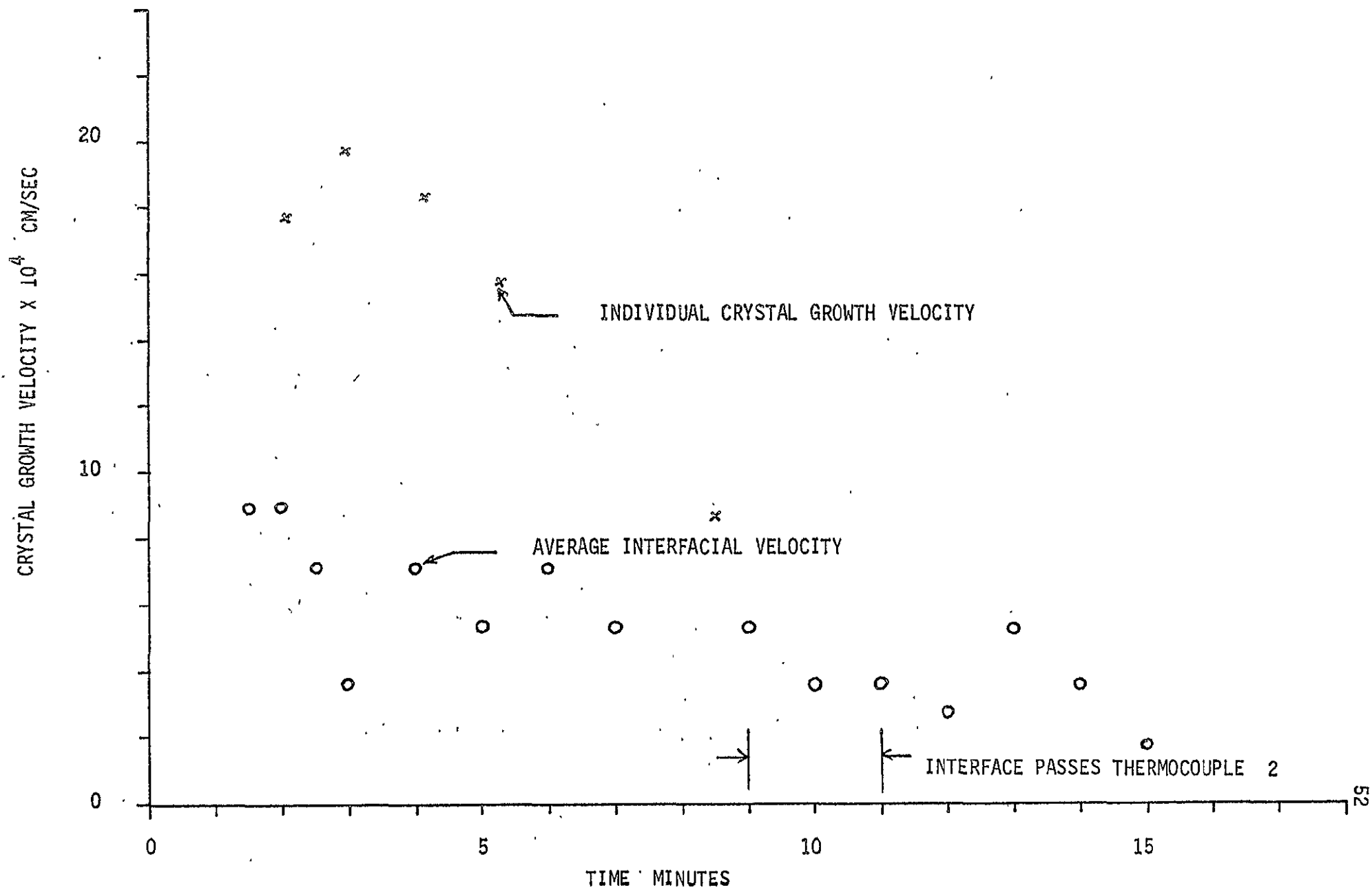


FIGURE 28
 RUN 6
 AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

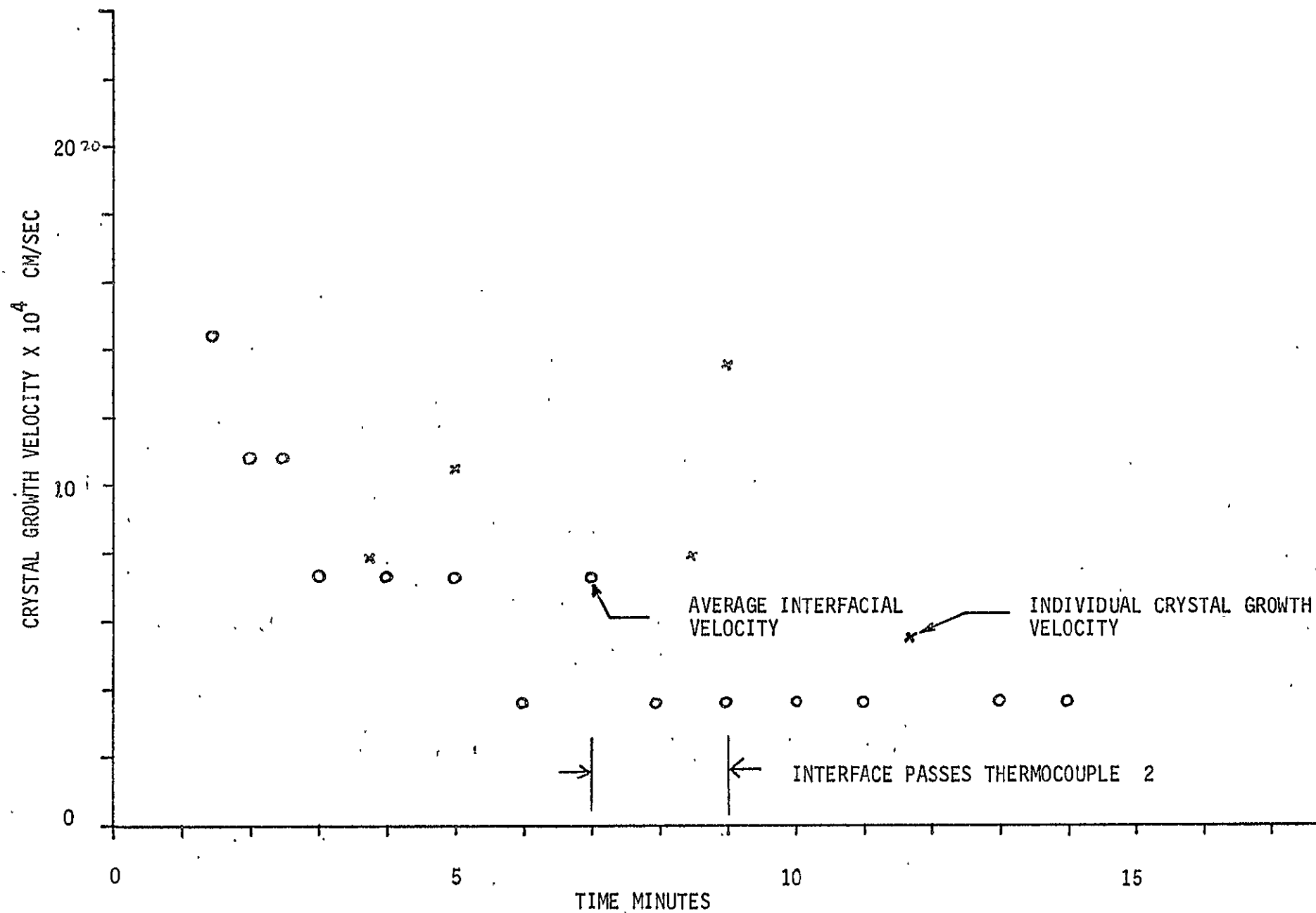


FIGURE 25³

RUN 7

AVERAGE INTERFACIAL VELOCITY AND INDIVIDUAL CRYSTAL GROWTH VELOCITIES VERSUS TIME

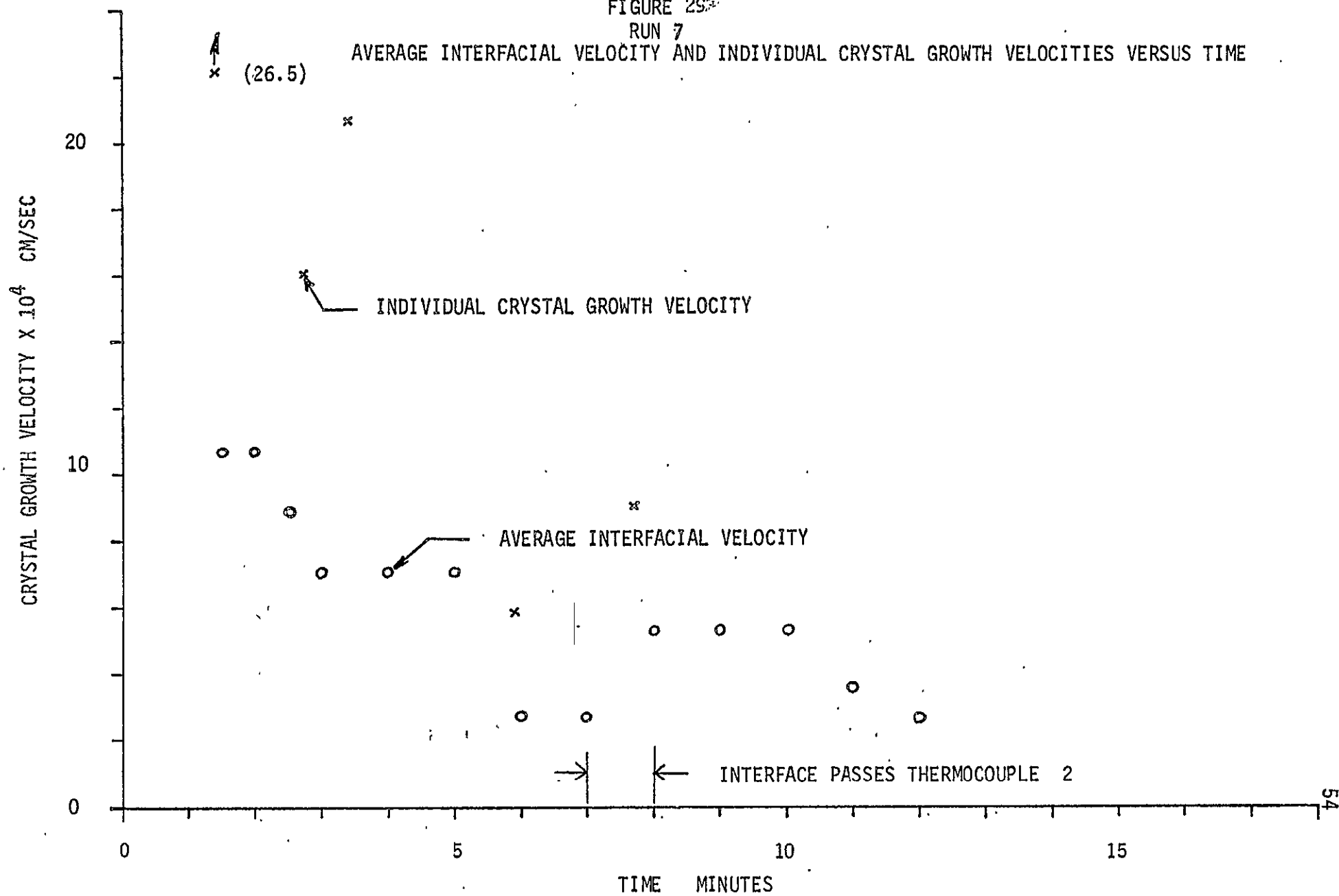
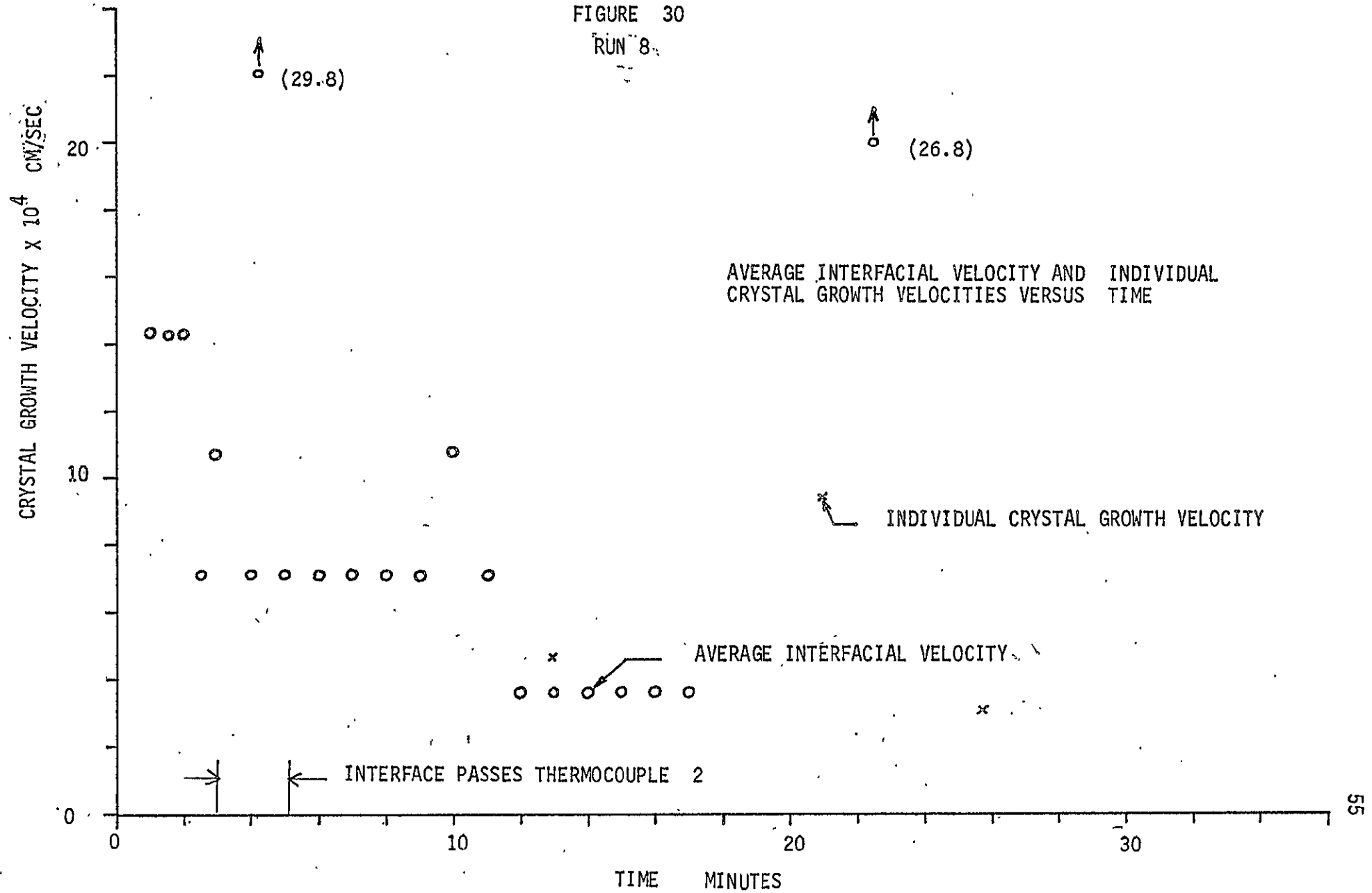


FIGURE 30

RUN 8




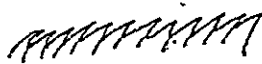



APPENDIX B

Optical Equipment List

| <u>Equipment</u> | <u>Catalog No.</u> <u>Van Waters & Rogers</u> |
|--|--|
| 1. EMDECO Drive for Bolex 16 | 47300-008 |
| 2. EMDECO Control for Camera Drive | 101-200 |
| 3. Focusing Eyepiece with Objective | 47-60-26 |
| 4. Monocular Photo Tube | 47-50-81 |
| 5. Stereo Microscope Body IV | 47-50-55 (less part 47-50-55-9001) |
| 6. Clamp on Stand D | 47-52-06 |
| 7. Eyepiece (10X) | 46-40-01 |
| 8. Large Connecting Ring | 47-60-02 |
| 9. Objective f = 100mm | 47-50-61 |
| 10. Adapter for Bolex 16 | 47-69-30 |
| 11. Basic Body I | 47-60-10 |
| 12. Large Connecting Ring | 47-60-02 |
| 13. Intermediate Piece with Rack and Pinion | 46-40-01 |
| 14. Stand with Circular Base | 46-72-80 |
| 15. Lamp Holder with Clamp, Filter Receptacle and Iris Diaphragm | |
| 16. Filter Holder | |
| 17. Adapter to use 12V Lamp | 46-70-42 |
| 18. Lamp Housing | 46-72-57 |
| 19. Bulb 12V, 60W | 38-02-16 |
| 20. Lamp Socket | 46-80-15 |
| 21. Transformer with Ammeter (regulating) | 39-25-63 |

APPENDIX C

Definition of Terms in the
Qualitative Observations

| <u>Term</u> | <u>Description</u> | <u>Sketch</u> |
|-------------|---|---|
| 1. Thorns | moderately broad, elongated crystals with either sharp or blunted tips |  |
| 2. Lawn | smooth interface, packed blades of grass, growth very even, growth uniformly curved |  |
| 3. Shrubs | long fat crystals, closely packed |  |
| 4. Bush | very irregular, closely packed and large crystals |  |
| 5. Mountain | large, massive crystals, pointed or flat tips |  |